TOPICAL REVIEW

Testing the limits of quantum mechanics: motivation, state of play, prospects

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Abstract
I present the motivation for experiments which attempt to generate, and verify
the existence of, quantum superpositions of two or more states which are by
some reasonable criterion 'macroscopically' distinct, and show that various
a priori objections to this programme made in the literature are flawed. I review
the extent to which such experiments currently exist in the areas of free-space
molecular diffraction, magnetic biomolecules, quantum optics and Josephson
devices, and sketch possible future lines of development of the programme.

1. Introduction

Quantum mechanics is very much more than just a 'theory'; it is a completely new way of
looking at the world, involving a change in paradigm perhaps more radical than any other in
the history of human thought. As we all know, quantum ideas were originally motivated by a
series of experimental anomalies at the level of single electrons, photons and atoms—atomic
structure, the photoelectric effect, the Davisson–Germer experiment and so on. And for many
years their main applications were at this or an only slightly more complicated level. As we go
into the 21st century, however, the considerations of quantum mechanics (hereafter QM) have
become increasingly indispensable tools for just about every sub-branch of physics, from the
cosmology of the early Universe to the psychophysics of human vision, from the structure of
the nucleon to the behaviour of the cuprate superconductors.

   Despite this enormous range of applications, however, there is a sense in which we can say
that the region of the whole parameter space over which the validity of QM has been directly
tested is still rather modest. Take, for example, the question of length scale. The majority
belief in the physics community would seem to be that the laws of physics as we currently
understand them hold at all length scales down to the Planck scale ($\sim 10^{-35}$ m) and up to the size
or 'characteristic scale' of the Universe ($\sim 10^{+30}$ m)—that is, over ~ 65 orders of magnitude.
Now it is certainly true that essentially quantum mechanical considerations have been applied
to explain phenomena occurring (or believed to occur) over almost all of this range, but in
many cases (and particularly at the upper end of the range) the regime in which QM needs
to be valid for the explanation to work is only the atomic and perhaps the 'near-subatomic'

regime. (For example, the phenomenon of hydrogen recombination, which is so essential a feature of the history of the early Universe in the standard model, is quintessentially quantum mechanical in nature, but clearly does not require that quantum mechanical interference can take place over a cosmic scale of distance!) In fact, if one were to ask over what length scales QM has been directly tested (in the sense of detection of characteristically QM effects such as interference) the answer would probably be: down to perhaps $\sim 10^{-18}$ m (in high-energy diffraction experiments) and up to a few metres or (recently) a few kilometres (in EPR-type experiments; see below)—that is, over $<30\%$ of the total (logarithmic) range over which most people believe it to be valid. Similar remarks apply to scales of (e.g.) time and, even more, mass.

Now, it is certainly true that over this still quite substantial slice of the whole parameter space there is satisfactory and often excellent evidence that the QM framework (of course when supplemented by more specific considerations) is quantitatively valid; and, equally important, that there is, at least at present, no positive experimental evidence that it is not valid in other regions where it has not been directly tested. In these circumstances the principle of Occam's razor would certainly suggest that the intellectually economical attitude is to assume that the general conceptual scheme embodied in QM is in fact valid for the whole of the physical universe without restriction; and this is indeed the attitude of the great majority of practising physicists at the dawn of the 21st century. In fact, to make a challenge to this view look other than 'scepticism for scepticism's sake', it would seem one should have some specific *a priori* reason for believing that QM may break down when pushed far enough along one or more of the 'axes' of the parameter space. Do such *a priori* reasons exist, and if so, what are the relevant axes?

I believe that at present we know of three principal 'directions' in which it is not unreasonable to look for a possible breakdown of QM. The first is the direction of very small length scales (which, to the extent that we stay within the relativistic quantum mechanical paradigm itself, is essentially equivalent to the direction of very short times and of high energies). As is well known, as one approaches the Planck scale ($\sim 10^{-35}$ m) the quantum fluctuations of the space-time metric which are predicted by the simplest ways of combining QM and general relativity become comparable to the mean value; so, if length scales shorter than this should turn out even to be meaningful, one might reasonably speculate that a description below this level might require fundamentally non-quantum mechanical concepts. Any such speculations are at present very far removed from direct experimental test, and I will not discuss them further here.

The second direction in which it would not, *a priori*, be unreasonable to conjecture a failure of QM is defined by the combination of (relatively) short times and (relatively) long distances, or more precisely, by the condition of space-like separation in the sense of special relativity. As is by now very widely known, in an epoch-making 1964 paper the late John Bell demonstrated that under such conditions the two-particle correlations predicted by QM are incompatible with a conjunction of very innocuous and commonsensical-looking postulates which nowadays are usually lumped together under the definition of an 'objective local' theory; crudely speaking, this class of theories preserves the fundamental postulates of local causality in the sense of special relativity and a conventional concept of the 'arrow' of time, and in addition makes the apparently 'obvious' assumption that a spatially isolated system can be given a description in its own right. The intuitive plausibility (to many people) of the class of objective local theories is so high that once Bell had demonstrated that under suitable conditions (including the condition of space-like separation) no theory of this class can give experimental predictions which coincide with those made by QM, a number of people, including some very distinguished thinkers, committed themselves publicly to the opinion that it would be QM rather than the objective local postulates which would fail under these anomalous conditions. Unfortunately

for these sceptics, over the last 30 years a series of experiments of increasing ambitiousness[1] have been made under just these conditions, and while there is still some argument (because of a couple of technical loopholes) about whether the outcome of these experiments excludes the class of objective local theories (a large majority of the physics community feels that it does), there is little argument that it is entirely consistent with the validity of the QM predictions. In other words, while the experiments do not exclude a possible failure of QM under conditions of space-like separation (if one takes a falsificationist point of view, no experiment ever could!), they certainly give no evidence whatsoever that such a failure is occurring. I shall not discuss this line of research (which will certainly continue to be vigorously pursued) explicitly any further in this review; for an excellent review at a fairly non-technical level, see [2]. However, it turns out that a number of technical questions which have arisen in this connection have close analogues in the material below, and I shall try to cite references to them at appropriate points.

The third 'axis' along which it is (at least in the opinion of the present author) not unreasonable to seek evidence of a breakdown of the quantum mechanical scheme of the physical world is that which connects the world of atoms and electrons, for which it was originally developed, with the 'everyday' world of our immediate experience. For the rest of this review, the phrase 'testing the limits of QM' should be understood to refer exclusively to tests in this direction. Exactly how to characterize the relevant 'axis' (or axes) is itself the subject of debate, and I will return to this question below. First, however, let us examine why this is a promising direction in which to look.

2. Motivation

Sixty-five years ago one of the founding fathers of QM, Erwin Schrödinger, put his finger on the problem that has troubled the physics community (or at least a part of it) ever since: if we assume that the formalism of QM can be extrapolated without essential change from the level of single electrons and atoms up to that of everyday life, we risk arriving at a description of the latter which is, at least *prima facie*, in violent conflict with our common-sense notions. In Schrödinger's original 'quite absurd' thought-experiment, a superposition of microscopically distinct states (the decayed and undecayed states of a radioactive nucleus) leads inexorably to a superposition of macroscopically distinct states (states in which a cat in a closed box is respectively dead and alive). As Schrödinger himself points out in the final, rather obscure sentence[2] of his paper [3], this description is not, conceptually, equivalent to a classical 'mixture' of the two states in question, that is, a description in which each individual cat in the relevant ensemble is either dead or alive, but we do not know without explicit inspection which.

Let us try to make the considerations underlying Schrödinger's argument more explicit. Imagine an ensemble of microscopic systems, e.g. neutrons, which can proceed from an initial state A via intermediate states B or C to a final state E (or alternatively to other possible final states D, F, . . . , but these will not be of explicit interest to us). The states in question might correspond to different spatial locations, as in a typical Young's-slits experiment, but could equally well correspond to a more abstract difference (as in the case, e.g., of different states of the neutral K-meson system with respect to its internal (strangeness/CP) degree of freedom). Let $P_{A \to B \to E}$ be the probability that a system originating at A arrives at E when the intermediate state B is the only one open, e.g. because we have physically obstructed the path via C; similarly, let $P_{A \to C \to E}$ be the probability of proceeding from A via C to E, when only C is open. Further, let us adopt the (possibly question-begging) notation $P_{A \to (B or C) \to E}$ for the probability that a system originating at A proceeds to E when the paths through both intermediate states, B and

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[1] See for example [1].
[2] '(There is) a difference between a blurred or unfocused photograph and a shot of clouds and mist'.

C, are open. Then, as we all know, it is a surprising but well-established experimental fact that under appropriate conditions we find the result

$$P_{A \to (B \,or\, C) \to E} - (P_{A \to B \to E} + P_{A \to C \to E}) \equiv K \neq 0 \tag{2.1}$$

where the 'interference' term K may, depending on the conditions of the experiment, be either positive or negative (and in extreme cases may cancel completely the second term on the LHS). In words, the total probability of arriving at E when both paths are open is not in general the sum of the probabilities of arriving through each path separately. Equation (2.1) is simply a statement of the experimental facts and in itself carries no particular theoretical implications.

Needless to say, the analysis of the above thought-experiment in standard quantum mechanical terms is very straightforward. We assign to each of the paths A $\to$ B $\to$ E a probability amplitude $A_{A \to B \to E}$ etc, and postulate that the total amplitude for arriving at E when both paths are open is the sum of the amplitudes for the different paths; moreover, we assert that the total probability of arrival at E under given conditions is the squared modulus of the amplitude for doing so. Then, omitting the couple of lines of simple algebra involved, we indeed find a result of the form (2.1), with the interference term K given by the expression

$$K = 2 \operatorname{Re} A^*_{A \to B \to E} A_{A \to C \to E}. \tag{2.2}$$

Equation (2.2) is a crucial ingredient in any discussion of the possible interpretations of the formalism of QM. Consider a given ensemble for which K is known, experimentally, to be non-zero. Equation (2.2) implies that for this ensemble the quantities $A_{A \to B \to E}$ and $A_{A \to C \to E}$ must be simultaneously non-zero. To make any positive assertion which goes beyond this, it seems that one needs to decide on the ontological status of the amplitudes $A_{A \to B \to E}$ etc. If these quantities characterize the ensemble as a whole and are merely auxiliary quantities whose only role is to permit the calculation of predicted possibilities (as is roughly speaking the attitude of the full-blooded 'statistical' interpretation; see below), then it seems nothing much further can be said, and in particular no positive conclusions can be drawn about the behaviour of individual particles of the ensemble. If, on the other hand, one is inclined to regard the amplitudes as corresponding to something 'out there' in the real world, then it would seem very difficult to avoid associating that something not just with the ensemble as a whole, but with the individual particles composing it, and it is then very tempting to draw the conclusion that each individual particle must in some sense 'sample' both of the apparently alternative paths simultaneously. It then of course becomes a problem to reconcile this conclusion with the well-known observation that any (effective) attempt to 'measure' which path an individual particle took yields a definite answer, i.e. each particle appears to take either the path via B or that via C. (This observation does not give rise to a problem in the statistical interpretation, since there the amplitudes correspond to nothing in the physical world.)

In the context of the present discussion, however, it is not particularly important to resolve the question examined in the last paragraph. What is important is that even in the absence of a resolution we can draw from equation (2.2) a negative conclusion. That is, the one interpretation of the QM formalism which we cannot give (or at least cannot give without extreme intellectual contortions; see below) is that each particle of the ensemble in question takes, even in the absence of 'measurement', either path B or path C, and the QM description is simply an expression of our ignorance as to which path was followed in a particular case. Were we to take this view, it would seem *prima facie* impossible to permit $A_{A \to C \to E}$ to be non-zero for a particle[3] which in fact took path B, and vice versa; so since each particle definitely took either path B or path C, for each particle one or other of $A_{A \to B \to E}$ and $A_{A \to C \to E}$ must be zero.

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[3] It is assumed, of course, in this interpretation that the amplitudes are associated with individual particles and not just with the ensemble as a whole.

The interference term is then automatically zero for each particle of the ensemble, and therefore for the ensemble as a whole, in contradiction to experiment. The only way around this argument would appear to be to deny the premise, i.e. to assert that even though a given particle 'actually took' route B, the associated amplitude for the path via C is nevertheless non-zero and has a physical effect. This is indeed in effect the assertion made in the Bohm interpretation of the QM formalism (see e.g. [4]). No experimental consequences are drawn from this assertion other than the standard predictions of the QM formalism, so whether one regards it as a substantive resolution of the apparent paradox or as little more than a reformulation of it is no doubt a matter of personal taste (the present author inclines towards the latter point of view). In any case, adoption of the Bohm viewpoint does not seem to me to affect the argument below in any substantial way, so purely for the sake of conciseness of presentation I will assume from now on that we do not adopt it and are therefore content to draw the negative conclusion above, namely that when an ensemble of microscopic entities is described in the QM formalism in the absence of 'observation' by a superposition of two (or more) non-vanishing probability amplitudes corresponding to (microscopically) different paths or 'outcomes', it is not the case that each entity of the ensemble definitely realizes one outcome or the other.

Before proceeding, let me make one thing quite explicit: in the above argument, we have drawn a (negative) conclusion about the *meaning* (or rather possible meanings) of the formalism of QM. The *evidence* for this conclusion is the finite value of the quantity K of equation (2.2), as measured experimentally in various atomic-level experiments. The truth or not of the conclusion is, as a matter of logic, independent of the evidence for it. As we shall see below, neglect of this apparently trivial consideration has led to a vast amount of avoidable confusion in the problem of quantum measurement.

While this conclusion looks at first sight strange and perhaps disturbing, three generations of physicists have by now been persuaded to take it in their stride. They have probably been thus persuaded, more than anything else, by the arguments of many of the founding fathers of QM, par excellence Nils Bohr, to the effect that microscopic entities such as electrons, photons and atoms 'exist' (if that is the right word) at a level so different from that of everyday experience that we should not expect our common-sense intuitions to apply to them. In particular, in Bohr's often-repeated view, electrons and atoms are simply not the kind of things which can be said to have definite properties in the absence of a specification of the macroscopic apparatus which we have put in place to examine them. Thus, in the example above (interpreting it for definiteness as a standard Young's-slits experiment with electrons) an electron passing through the relevant set of slits etc in the absence of a macroscopic device specifically set up to detect its passage is simply not the same object as an electron passing through the same device but subjected to a 'which-slit' ('which-path') measurement, and consequently no genuinely paradoxical conclusions can be drawn. In a similar spirit, some empiricist philosophers such as Hans Reichenbach [5] have argued that the very ontological status of microscopic entities is itself a matter of convention, and thus (in effect) that the whole scheme of QM is nothing but a formal calculus, designed purely to predict the probabilities of various outcomes at the everyday, macroscopic level, which we need feel no inescapable duty to interpret at all. The approaches of Bohr and Reichenbach[4] are, of course, both variants of the general approach to the formalism of QM which goes under the name of the 'Copenhagen interpretation' (though it should probably more correctly be called the 'Copenhagen non-interpretation', since its whole point is that any attempt to interpret the formalism in intuitive terms is doomed to failure). I would regard it as a defining component of this interpretation, which distinguishes it from the 'statistical' interpretation (on which more below) that while denying that microscopic

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[4] With my definition of 'Copenhagen' as opposed to 'statistical', Reichenbach in his book actually considers both possibilities.

objects (electrons, photons, atoms, ...) necessarily have definite properties in the absence of observation, it emphatically asserts (or at least implies) that macroscopic objects (counters, cats etc) do have such properties at all times, whether they are observed or not. This insistence on the necessity of drawing a sharp line (even though its actual position may be somewhat arbitrary) between the 'microscopic' world of electrons and atoms and the 'macroscopic' one of everyday life (including measuring apparatus) is a pervasive theme in the writings of Nils Bohr, and is precisely the point of attack of Schrödinger's 1935 argument, to which I now turn.

An unspoken but crucial assumption which underlies Schrödinger's argument, and the many more recent variants of it, is the following: since quantum mechanics, as is universally agreed, describes brilliantly the behaviour of individual electrons, atoms etc, and since (as again everyone agrees) everyday objects such as cats and counters are composed of electrons and atoms, it therefore follows that QM in principle describes the behaviour of these everyday objects just as perfectly as it does that of electrons etc. This of course is not to say that it is always necessary to give a QM description of everyday objects; as we all know, under most circumstances classical physics gives an excellent account of their behaviour, at least as regards crude properties such as the centre-of-mass dynamics. However, for all conceivable properties and behaviour (including that well described by classical physics) it should in principle be legitimate to ask for a quantum mechanical account, which in particular will respect the linear character of state evolution (see below). Only a small minority of physicists (which needless to say includes the present author!) would regard this assumption as other than totally obvious, so let us for the moment accept it and see where it leads.

Where it leads, of course, is to the notorious 'cat' (or measurement) paradox of Schrödinger's original paper. It is quite easy to conceive experimental set-ups (Schrödinger's original proposal is just one particularly dramatic special case) in which one microscopic state—call it ψ_1—leads to a particular macroscopic outcome, say '1', while a different microscopic state ψ_2, orthogonal to ψ_1, leads to a macroscopically different outcome, say '2'; this is an experimental fact which can be checked without any particular theoretical preconceptions (for example, set up an arrangement similar to that of Schrödinger's original paper, whereby the decay of a radioactive nucleus triggers a counter which in turn triggers a gun etc[5]). How do we describe this state of affairs in the language of QM? In real life it is very unlikely that we know enough details of the initial state of the macroscopic body (cat, counter, ...) in question to assign to it a pure quantum mechanical state, and we should realistically have to describe it by a density matrix. However, this consideration simply complicates the notation without in any way blunting the force of the paradox (see e.g. [6]), so purely for convenience of presentation I shall postulate, of course unrealistically, that the initial state of the macroscopic object (and any parts of the universe which are correlated with it) is a pure QM state, say Ψ_0, and thus the combined system formed by the microscopic and macroscopic objects is described by the product $\psi_1\Psi_0 \equiv \chi_1$ (or $\psi_2\Psi_0 \equiv \chi_2$).

To describe the situation in quantum mechanical language, we now require that the time evolution operator $\hat{U}(t)$, which must couple the microscopic and macroscopic systems, has the properties

$$\hat{U}(\infty)\chi_1 \equiv \hat{U}(\infty)\psi_1\Psi_0 = \psi_1'\Psi_1 \equiv \chi_1' \tag{2.3}$$

$$\hat{U}(\infty)\chi_2 \equiv \hat{U}(\infty)\psi_2\Psi_0 = \psi_2'\Psi_2 \equiv \chi_2'. \tag{2.4}$$

We need not be concerned in this context with the final states ψ_1', ψ_2' of the microsystem (which need not even necessarily be mutually orthogonal); what concerns us is the final states Ψ_1, Ψ_2 of the macroscopic system. Since these are supposed to correspond to the outcomes '1' and

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[5] In principle we could perform an independent check that the nucleus has in fact decayed, e.g., by detecting the recoiling daughter nucleus (assuming that the counter is triggered by (say) an emitted α-particle).

'2' above, the two quantum states in question must represent macroscopically distinct states of the universe (e.g. 'cat alive' and 'cat dead') and thus must *a fortiori* be mutually orthogonal.

To generate a paradox, we imagine that we start our ensemble of nuclei (or whatever) in a linear quantum superposition of the two microscopically distinct states ψ_1 and ψ_2 (in many implementations of practical interest there is no particular difficulty in doing this). The initial state of the combined microsystems and macrosystems is then

$$\chi = (a\psi_1 + b\psi_2)\Psi_0 \equiv a\chi_1 + b\chi_2 \qquad |a|^2 + |b|^2 = 1. \tag{2.5}$$

The crux, now, is that the evolution operator $\hat{U}(t)$ is strictly linear (a very fundamental and non-negotiable ingredient of the QM formalism), and thus equations (2.3) and (2.4) rigorously imply the result

$$\hat{U}(\infty)\chi = a\chi_1' + b\chi_2' \equiv a\psi_1'\Psi_1 + b\psi_2'\Psi_2. \tag{2.6}$$

Thus the final state of the universe (or more strictly ensemble of universes) corresponds to a quantum superposition of macroscopically distinct states—in Schrödinger's original example, states in which the cat (assumed to be in a closed box and thus unobserved) is respectively dead and alive. Nevertheless, whenever experiments of this type are actually done and we 'open the box'—if not with cats, at least with macroscopic counters etc—it is our direct experience that on each particular run a definite macroscopic outcome always occurs: either the counter clicks or it does not, the monitor reads 5 or 10 A, not both simultaneously[6] etc.

To try to catalogue here the complete range of 'solutions' of the quantum measurement paradox which have appeared in the literature since Schrödinger's original paper would be both impractical and pointless. Broadly speaking, however, I believe that the vast majority of such 'solutions' can be grouped into three categories[7], depending on the status which they attribute to the probability amplitudes appearing in the QM formalism at the microscopic and macroscopic levels. At one extreme one has one version or other of the 'statistical' interpretation; for a trenchant defence of this approach, see e.g. [8]. This interpretation starts off from considerations similar to those of the classic 'Copenhagen' approach, but extends them to their logical conclusion; thus, it refuses to attribute any kind of 'reality' to the QM probability amplitudes either at the microscopic or, *a fortiori*, at the macroscopic level. According to this view, the QM amplitudes correspond to nothing whatsoever in the physical world, but are simply intermediate symbols in a calculus whose only ultimate function is to predict the statistical probability of various directly observed macroscopic outcomes, and no further significance should be attributed to them. For an advocate of this class of solution (at least according to the present author's understanding) any questions about the 'true' state, not just of an electron in a Young's-slits experiment but also of an unobserved cat in the Schrödinger thought-experiment, are strictly meaningless; the only kind of question which can be meaningfully asked is 'what is the probability that, if I open the box containing this particular cat, she will be found to be alive?'—a question to which, of course, the formalism of QM when augmented with the standard measurement axioms always gives a well-defined answer.

At the opposite extreme to the statistical interpretation we find the 'relative-state' interpretation of Everett [9], often known colloquially, since its popularization [10] by de Witt, as the 'many-worlds' (or 'many-universes') interpretation. According to this view, the quantum mechanical probability amplitudes reflect physical reality, both at the microscopic level and at the macroscopic level. Thus, in Schrödinger's thought-experiment, each individual cat of the ensemble is in some sense simultaneously dead and alive, and remains so even when we

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[6] Indeed, it is not easy to imagine what it would be like to 'experience' a quantum superposition of the type (2.5).

[7] A few solutions, such as that proposed by Schulman [7], may not naturally fall under this categorization. I do not include here resolutions of the paradox which rely on a breakdown at some point of the QM formalism; see below.

open the box and inspect her; my consciousness that I see a particular cat to be (say) dead is an illusion, which can be consistently maintained only because of formal theorems of QM which guarantee that the amplitude for me to see this particular cat dead and for you to see her alive is zero. The branches of the superposition which we are not conscious of observing are said to be 'equally real' (though it is not clear to the present author, at least, what these words, ostensibly English, are suppose to mean).

In between the two above extremes there falls a vast class of variants of what I will loosely call the 'orthodox' interpretation of QM; I believe this name is justified by the fact that it seems to be the interpretation to which most physicists who actually use the formalism of QM in their everyday work tend to gravitate. Crudely speaking, advocates of this interpretation try to have their cake and eat it: to ascribe a sort of physical reality, at least in the negative sense, to the QM amplitudes at the microlevel but to deny it to such amplitudes at the macrolevel. That is, I believe a typical advocate of this kind of interpretation, if asked whether a particular electron in a Young's-slits experiment definitely goes through one slit or the other, would reply that it does not, and thereby implicitly agree that the question has a meaning; while the same advocate, if asked whether in Schrödinger's thought-experiment each individual cat is definitely alive or dead before any observation is made, would reply that she is. More generally, he/she would take the view that while at the microscopic level a quantum superposition indicates a lack of definiteness of outcome, at the macroscopic level a similar superposition can be interpreted as simply a measure of the probability of one outcome or the other, one of which is definitely realized for each individual number of the ensemble. The reason that such a dichotomy of interpretation is held to be legitimate is the phenomenon of 'decoherence' which will be discussed in detail in section 4: by the time we reach the level of cats or counters, any realistic QM calculation, which must take account of the influence of this environment, lack of knowledge of initial conditions etc, will automatically predict that there is no longer any possibility of observing interference between the two possible macroscopically distinct outcomes. 'Thus', it is argued, it is legitimate to take the view that one outcome or the other had by this stage definitely occurred in each case.

If regarded simply as a rule of thumb for deciding when one can in everyday practice afford to stop speaking the language of QM and replace it with the standard 'classical' description in which things do or do not happen etc (an 'for all practical purposes' ('FAPP') principle, in the language of the late John Bell), the above argument is unexceptionable. As a resolution of the philosophical paradox posed by Schrödinger, on the other hand, it is in the opinion of the present author (and incidentally also that of Bell [11]) a non-starter. At the microscopic level we adopted a particular interpretation of the formalism of QM, at least in a negative sense: a quantum superposition of two possibilities corresponds to a situation where it is not the case that each individual member of the ensemble realized one outcome or the other. The evidence for this interpretation was the phenomenon of interference, that is an (experimentally observed) non-zero value of K in equation (2.1). By the time we get up to the macroscopic level, it is agreed that at least under normal circumstances (see below) the evidence is no longer available, in the sense that the quantity analogous to K is now (experimentally observed to be) zero. However, the formalism of QM has in no way changed between the microlevels and macrolevels. If, therefore, one believes (as the overwhelming majority of physicists at present do) that the extrapolation of the formalism in this way is legitimate, then the attempt to exploit the vanishing of the interference term to justify a radical reinterpretation of the meaning of the QM formalism simply involves a logical fallacy: the meaning of the formalism is logically independent of the evidence that that understanding of the meaning is correct![8]

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[8] This argument is independent of the consideration, repeatedly emphasized by Bell, that one can never be 100% sure that K will in fact be zero in all possible experiments.

Thus, in the present author's view, of the three major classes of 'resolution' of the quantum measurement paradox, the 'orthodox' one involves a major logical fallacy and the 'many-worlds' interpretation is simply a meaningless collage of words. The 'statistical' interpretation, if taken to its logical conclusion, is internally consistent but conflicts rather violently with the 'realistic' intuitions which most practising physicists probably find not only philosophically congenial, but almost essential, psychologically, in their everyday work. Thus, one is led to consider the possibility that the fundamental premise of the argument is wrong: that is, that the linear formalism of QM does not apply in unmodified form to macroscopic systems in the same way as it does to their microscopic constituents. In the next section I consider this proposition, and in section 4 go on to whether it is experimentally testable.

3. The hypothesis of macroscopic realism

The suggestion that the formalism of QM as we know it at the atomic level might break down at some point along the road from there to the everyday world is by no means a novel one; indeed, it may be regarded as at least implicit in Schrödinger's original paper. In 2001, however, any concrete hypothesis along these lines must satisfy a rather severe set of constraints: not only must it (if it is to have any chance of being considered seriously by the vast majority of the physics community) respect the basic principles of special relativity and conventional notions about the arrow of time, it must also make experimental predictions which are consistent with those of standard QM over the substantial region of the parameter space where the latter has been tested and found satisfactory.

Of the various concrete schemes which have been proposed with a view to resolution of the measurement problem, by far the best-developed one is that associated with the names of Ghirardi, Rimini, Weber and Pearle (GRWP). I refer the reader to [12] or [13] for a detailed description of the GRWP theory and the current position with respect to experimental tests of it, but a very crude synopsis would go as follows: one postulates the existence of a universal background of a kind of noise, but one which is not itself describable in QM terms (this feature is absolutely crucial to the viability of the GRWP scheme). The effect of this universal noise background on physical objects, be they electrons or cats, is to add to the standard linear time-dependent Schrödinger equation an extra stochastic term which preserves neither linearity nor unitarity, and whose effect is, crudely speaking, to drive a quantum superposition of states (as defined in an appropriate basis; see below) into one of its branches; the detailed prescription for how this 'reduction' happens is constructed (as it must be, in order to agree with the predictions of standard QM) in such a way that if the original superposition was of the form $\sum_i c_i \psi_i$, the probability of ending up, after a sufficiently long time, in branch i is proportional to $|c_i|^2$. Evidently the prescription requires us to specify a 'preferred' basis; in the current version of the theory this is the position basis, so a superposition of very different position eigenstates will eventually be reduced while superpositions of different momentum eigenstates need not be. As currently constructed, the theory contains two adjustable parameters: a length scale (a) which determines the minimum difference in position between two or more branches which is necessary to trigger the reduction process, and a quantity (λ) which characterizes the 'efficiency' of this process, as measured by the rate at which a superposition state of a microscopic object such as an electron takes place. A very fundamental aspect of the GRWP theory, which is not built in by hand but is embedded in its structure, is that a superposition state of a complex body containing N correlated micro-objects (e.g. a U^{238} nucleus, with 92 electrons and 238 nucleons, so $N = 330$) is reduced at a rate which is of order $N\lambda$ rather than λ. Thus, for example, it is possible to choose λ to be small enough that superpositions

of photon states reaching us from the most distant quasars in the known universe are not appreciably reduced (so that Michelson interferometry still works!) while the final state of (each particular) cat in Schrödinger's thought-experiment is projected into either the dead or the living state in a time much shorter than could conceivably be resolved by human senses. In the current version of the theory the parameters a and λ are tentatively fixed at $\approx 10^{-5}$ cm and $\approx 10^{-16}$ s^{-1} respectively, and it seems that they could not be varied by many orders of magnitude from those values without leading either to a contradiction with the predictions of QM at the atomic level or to a failure to reduce macroscopic-level superpositions over typical 'human' timescales.

One aspect of the GRWP theory in its current formulation which may be relevant to the discussion below (though it is not clear that it is an indispensable ingredient) is the singling out of the centre-of-mass degree of freedom as 'special' in the context of the triggering of reduction: other kinds of difference between the branches of the superposition, e.g. difference in magnetic moment (cf below), do not as such trigger reduction.

While the GRWP theory has been most extensively developed, various other proposals have been made along somewhat similar lines; for example, Penrose [14] has suggested that collapse of a quantum superposition into one of its branches takes place as soon as the gravitational self-energy associated with the different mass distribution in the branches in question exceeds that of a single graviton. Below, I shall take a more heuristic approach to the measurement problem, in that I shall consider a whole class of theories or models which have in common only the property that quantum superpositions of macroscopically distinct states do not exist in nature; exactly at what stage and by what mechanism such superpositions are avoided is left open. Strictly speaking, with the technical definition of 'macroscopically distinct' which I suggest below, neither the GRWP theory nor the model proposed by Penrose falls into this class, since in each case some kinds of superposition survive, at least for a time, against reduction; however, I would regard this as an inessential feature of the model in question, and in any case, the general principle to be used below can be straightforwardly extended to such models. I will say that the class of theories which I consider below embody the principle of macroscopic realism (or macrorealism for short); clearly, both the GRWP and the Penrose models (and some others in the existing literature) are very much in the spirit of macrorealism, even if they do not conform to the letter of the preliminary definition above.

The provisional[9] definition of macrorealism used in the last paragraph involves the notion of two or more quantum states as being 'macroscopically distinct', and this phrase (or related ones) has caused an amount of controversy in the literature of the subject which seems to me quite disproportionate to any real points of physics which are at issue. My reason for using the phrase at all has simply been to try to capture our intuitive sense that a superposition of (e.g.) the living and dead states of a cat is much more of an affront to our common-sense notions about the world than, say, a superposition of position states (even if entangled with an internal degree of freedom; cf below) of a single atom[10]. What, exactly, is it that makes it so? (What is the correct measure of 'Schrödinger's-cattiness'?) Ideally, one would like a quantitative measure which corresponds to our intuitive sense; I shall attempt one below, but would emphasize that the choice between this and a number of similar and perhaps equally plausible definitions is, with one important exception (see below), very much a matter of personal taste, and that I very much doubt that 50 years from now anything of importance will be seen to have hung on it.

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[9] Later we will need a more technically precise definition: see section 6.

[10] Despite the fact that the words 'Schrödinger's cat' seem to be freely used in the atomic-physics and quantum-optics literature in connection with such states—a usage which seems to me a distinct stretch of language.

My own choice for the definition (or measure) of the degree of 'macroscopic distinctness' of two quantum states is actually a combination of two different quantitative measures. The first is straightforward: consider a particular extensive physical quantity i characterizing each of the states in question (e.g. total charge, total magnetic moment, total momentum, ...) and choose a 'reference' value of this quantity which is 'typical' at the atomic scale. (For example, in the case of magnetic moment the obvious reference value is the Bohr magneton.) Then express the difference in the expectation value of this quantity in the two states compared in units of the reference value, and call the result Λ_i. The maximum value of Λ_i over the various relevant extensive quantities i will be denoted simply as Λ and be called, when a name is necessary, the 'extensive difference' of the two states in question (or of their superposition).

Although the possession of a sufficiently large Λ is plausibly a necessary condition for two states of a superposition to be 'macroscopically distinct', it is less obvious that it is sufficient. To see the kind of problem involved, consider as a possible choice of i the quantity $\sum_\ell \boldsymbol{r}_\ell \equiv M\boldsymbol{R}$, where M is the total mass and $\boldsymbol{R}$ the position of the centre of mass. We might take the reference value in this case to be, say, a nucleon mass times a typical atomic dimension, let us say ~ 1 Å. Now consider a single neutron passing through a standard interferometer, with arms say 10 cm apart; we find that according to our definition the value of Λ_i is about 10^9! Yet I think most people would not want to say that the neutron interferometer is an example of a 'Schrödinger's-cat' situation. So we need some ingredient beyond mere extensive difference.

The second ingredient which I shall use as a measure of 'macroscopic distinctness' is a quantity which was first introduced in [15], and there named 'disconnectivity' and denoted as D, a notation I shall follow here. The intuitive content of this concept is essentially identical (at least when applied to pure states) to that of the idea of 'degree of entanglement' which has been intensively studied in the last few years in the area of quantum information: crudely speaking, a superposition of two or more N-particle quantum states possesses a high degree of disconnectivity (entanglement) if, in order to distinguish it from the corresponding classical mixture of the states in question, it is necessary to measure correlations of many ($\sim N$) particles. The question of a quantitative definition of D is quite tricky: a tentative definition was already given in [15], but from the point of view of modern quantum information theory it is no doubt inelegant and could almost certainly be substantially improved. Since we will be concerned, below, only with the order of magnitude of D and not with its exact value, the precise definition is of no great interest and there is no need to spend time on it here; when a concrete definition is needed, I will use that of [15] with the minor modification (2) below. However, several general points are worth making:

(1) With any reasonable definition of D, states of complex physical systems which have large values of the extensive difference Λ are likely also to correspond to values of D which are of the order of the total number of microscopic constituents. For example, the state at the intermediate screen of a Young's-slits apparatus of a U atom has a substantial value of Λ and also a value of D approximately equal to 330.

(2) A technical detail: in the case of a set of identical particles obeying Fermi or Bose statistics, we probably do not want the definition of D to include the correlations which are required purely by the necessity of (anti)symmetrizing the N-particle wavefunction. This point was not addressed in [15], but is easily accommodated by reformulating the definition in the language of second quantization.

(3) Under ordinary conditions, the typical states of so-called 'macroscopic quantum systems' (lasers, superconductors, superfluid He, ...) do not correspond to large values of D (in fact, a typical value of D for a laser or superfluid ^{4}He is about 1 and for a superconductor 2!) This point, which is exhaustively discussed in [15], unfortunately appears still to be misunderstood in some of the quantum measurement literature.

(4) Finally, it is essential to appreciate that the definition of D is unique only to the extent that we implicitly specify the appropriate quantum mechanical basis, namely the basis of products of single-particle wavefunctions. D is invariant against alternative choices of the single-particle basis, but it is emphatically not invariant against e.g. the transformation to centre-of-mass ($\boldsymbol{R}$) and relative (ρ) coordinates[11] (in the latter basis, since the wavefunction of the U atom at the intermediate screen is schematically $(a\chi_1(\boldsymbol{R}) + b\chi_2(\boldsymbol{R}))\psi(\rho)$, the value of '$D$' would be 1!) That the system should, as it were, care whether we use the single-particle basis or the COM/relative basis appears of course within the framework of QM very strange, since it is part of the beauty of QM that all possible bases have equivalent status; however, the whole point of introducing D is the hope that when it reaches some large value some currently unknown theory alternative to QM may take over—and that alternative, non-quantum mechanical theory may indeed[12] 'care'!

My definition of the degree of 'macroscopic distinctness' of the two (or more) states of a superposition will thus combine in one fashion or another (e.g. by multiplying them) the relevant values of the 'extensive difference' Λ and the 'disconnectivity' D, and I will characterize various existing and contemplated experiments by the values of these two measures occurring in them. I emphasize again that there is nothing sacrosanct about this particular definition, and that many alternative formulations are possible. However, there is one definition of 'macroscopically distinct' that has unfortunately appeared on more than one occasion in the quantum measurement literature which I believe to be distinctly unhelpful, at least in the present context. Namely, it is sometimes argued that two quantum states can only be legitimately said to be 'macroscopically' distinct if there is no possibility of observing the effects of interference between them. Note only is this criterion inherently ambiguous (does 'no possibility' refer to the present, to 100 years from now or what?), but even if it could be made foolproof, such a definition would of course make any experimental programme such as that outlined in the next section pointless by construction. In the present author's opinion, there is little to be gained by using purely verbal manoeuvers to dismiss physically interesting questions.

Once we have a definition of 'macroscopically distinct', be it the above one or some (reasonable) alternative, we can ask the question: is it possible to determine experimentally whether superpositions of macroscopically distinct states actually occur in nature, or whether, rather, one state or other is always definitely realized (the hypothesis of macrorealism)? It is to this question that I turn in the next section.

4. Experimental tests: general considerations

Let us suppose we take the view that the quantum measurement paradox is sufficiently worrying that it is not unreasonable to consider seriously the generic hypothesis of macrorealism. Is there then any possibility of doing relevant experimental tests, or is the question destined to remain forever 'philosophical' (a term which in the loose usage of physicists seems to denote just about any issue to which they can currently foresee no experimental input)?

[11] Of course we could make a similar point with respect to the concept of 'entanglement' as discussed in quantum information theory: e.g. with the usual (single-particle) choice of basis the $S = 0$ state of two spin-1/2 particles is indeed maximally entangled, but if we were to work in the basis defined by S and S_z it would of course not be entangled at all. Needless to say, in the context of discussions of non-locality etc, the single-particle basis is overwhelmingly 'natural', which is no doubt why this point is not much discussed.

[12] We may compare the situation with respect to a natural 'scale' of the product of energy and time (cf [16]): within the framework of classical mechanics any such scale would be highly unnatural, yet it is of course fundamental in QM.

A very well-worn theme in the discussion of the paradox over the last 60 years has been that once one's quantum superposition has progressed to the macroscopic level, so that formally the description of the universe (ensemble of universes) is of the schematic form $c_1\Psi_1 + c_2\Psi_2$, where Ψ_1 and Ψ_2 represent macroscopically distinct states, then no possible measurement can show the effects of interference between the two states. In other words, a correct application of the QM formalism will give, for all possible experiments, precisely the same predictions as would follow from the assumption that by this stage each individual system of the ensemble has realized either state Ψ_1 or state Ψ_2, with probability $|c_1|^2$ or $|c_2|^2$ respectively. The reasons for this widespread opinion, which have to do with the phenomenon of decoherence and related things, will be explored further below; at this point we merely note that if it is true, it precludes forever any possibility of discriminating experimentally between the hypothesis that the formalism of QM remains valid at the macroscopic level and the alternative hypothesis of macrorealism, since any remotely plausible macrorealistic model will have to produce, *inter alia*, the result that the probability of realization of macrostate is $|c_i|^2$ and thus will make predictions identical to those which are made (under the above assumption) by standard QM. To put the same point in a more positive way, a necessary (though, as we shall see, not by itself a sufficient) condition for being able to discriminate experimentally between QM and macrorealism is that, in the experimental situation in question, application of the standard QM formalism leads to the prediction of non-zero effects arising from the quantum interference between macroscopically distinct states (hereafter abbreviated QIMDS).

Why is the simple observation of QIMDS at a given level insufficient to discriminate between QM and macrorealism at this level? For a simple logical reason: if theory T unambiguously predicts experimental consequence E, then the observation of not-E certainly implies the falsity of T, but it is a fallacy (called by logicians 'affirmation of the consequent') to maintain[13] that observation of E establishes the truth of T. Thus, the observation of QIMDS does not, logically, establish the truth of QM, and it establishes the falsity of macrorealism only to the extent that the latter has been explicitly shown to predict the absence of QIMDS. At the time of writing, this has been done only for a certain specific type of experiment (described below), not for the whole range of QIMDS-type phenomena (and in particular not for the types of experiment conducted to date; see section 5).

From a methodological point of view, therefore, any progress in discriminating between QM and macrorealism may be conceived of having three stages (of which one or more may turn out to be unnecessary in practice for a specific type of experimental system).

Stage 1. One conducts circumstantial tests to check whether the relevant macroscopic variable appears to be obeying the prescriptions of QM.

Stage 2. One looks for direct evidence for QIMDS, in contexts where it does not (necessarily) exclude macrorealism.

Stage 3. One conducts an experiment which is explicitly designed so that if the results predicted by QM are observed, macrorealism is thereby excluded;

I discuss the nature of one such possible experiment in section 6.

When about twenty years ago such a programme began to be consciously envisaged, it ran into a gamut of objections, which (probably not untypically!) ranged from the claim that it was trivial (i.e. superfluous) to the position that it was ridiculous (i.e. impossible). I believe that the claims of superfluity were (and, in so far as they are still current, are) based on

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[13] Of course the physics literature is chock-full of papers which formally affirm the consequent ('our experimental result E proves that theory T is correct'). In such cases there is usually an unstated (no doubt sometimes unconscious!) premise, namely 'E is so unexpected/spectacular that any theory T$'$ other than T would have predicted not-E'.

misconceptions concerning what exactly is demonstrated by previously existing experiments. This topic is discussed in some detail in [15], and I will comment here only on two specific misconceptions which seem to be particularly persistent. First, it is sometimes claimed that the experiments conducted since 1969 in connection with Bell's theorem are already sufficient proof that macrorealism is false. This is simply incorrect: in fact, it is easy to see that any 'actualization' of a particular state of the macroscopic counters (or for that matter of the photons themselves, once they have passed or failed to pass the relevant polarizer!), even at a stage well before the final 'observation', would in no way change the results predicted by a complete QM treatment[14]. Secondly, one not infrequently hears the view expressed that even experiments on 'simple' (microscopic) quantum systems, while they do not in general involve large values of the measure I have called 'extensive difference', do nevertheless often involve high degrees of disconnectivity (entanglement). A typical example which is cited in this context is the superposition of the vacuum and one-photon states of the electromagnetic field in a high-Q microwave cavity; it is argued that the mere fact that the photon is confined within the cavity already shows that it must interact strongly with the electrons in the walls of the latter, and that this in turn implies that the quantum state of the electromagnetic field is strongly entangled with that of electrons. Again, this argument is simply incorrect; a straightforward QM calculation shows that the deviation of the disconnectivity from its trivial value unity is by any reasonable definition only of the order of (some power of) the inverse of the cavity Q-factor, i.e. very tiny[15] (typically in such cases Q is of order 10^9).

More worrying than objections that the experimental programme is superfluous has been, at least historically, the opinion that it is unrealistic to the point of impossibility. The argument to this conclusion comes in a number of variants, but most of them boil down in the last resort to invocation of the phenomenon of 'decoherence'. Although the name is fairly recent, the importance of this phenomenon in the context of the quantum measurement problem has been recognized for many decades (see e.g. [18]); for a readable review, see [19], and for a more technical discussion [20]. It is probably most easily formulated within the language of macroscopic variables. Consider, for example, a Young's-slits-type state of the centre of mass of a complex object (say a golf ball) of the form

$$\Psi = a\Psi_1(\boldsymbol{R}) + b\Psi_2(\boldsymbol{R}) \tag{4.1}$$

where $\Psi_1(\boldsymbol{R})$ and $\Psi_2(\boldsymbol{R})$ correspond to localization of the COM position $\boldsymbol{R}$ around two different points $\boldsymbol{R}_1$ and $\boldsymbol{R}_2$ which are 'macroscopically' separated (say $\sim$10 cm). As we shall see in section 5, under suitable circumstances this wavefunction, if correct as assumed, will give rise to QIMDS, as evidenced e.g. by an interference pattern on a final screen. The 'decoherence' argument now points out that there will be any number of degrees of freedom of the 'environment' (a blanket term meaning in effect anything outside the degree of freedom of interest which interacts with it[16]) which are sensitive to the state of the system and will therefore become correlated with it. (For example, in the case of the golf ball the state of the radiation field will be affected differently by a ball at $\boldsymbol{R}_1$ and one at $\boldsymbol{R}_2$; cf [20].) Thus, if we denote the degrees of freedom of the environment schematically by ρ, the current description of the 'universe' ($\equiv$system + environment) is by an entangled wavefunction of the form

$$\Psi_u = a\chi_1(\rho)\Psi_1(\boldsymbol{R}) + b\chi_2(\rho)\Psi_2(\boldsymbol{R}) \tag{4.2}$$

[14] As Stapp [17] has shown, it is possible to reformulate the class of 'objective local' theories which are (usually believed to be) refuted by the experiments in terms of a postulate of (macroscopic) 'counter-factual definiteness'. However, this is quite different from macrorealism.

[15] Indeed, if we really had $D \gg 1$ as claimed, it would be paradoxical that the experiments are so well described by the naive textbook QM of an 'isolated' system!

[16] Note that if e.g. the principal degree of freedom is the centre-of-mass position of a body, the 'environment' may include its internal coordinates (though see below).

where the states χ_1, χ_2 of the environment are to a high degree of approximation mutually orthogonal, and it is then impossible (or so it is claimed) to obtain the phenomenon of QIMDS, since in the context of any measurement on the system (uncorrelated with a measurement on the environment) the wavefunction (4.2) is equivalent to a classical mixture of the two branches with weights $|a|^2$ and $|b|^2$ respectively. We can reformulate the argument in the language of density matrices if we consider the reduced density matrix $\hat{\rho}_s$ of the system, that is the density matrix of the universe traced over the environmental degrees of freedom; then 'before decoherence', i.e. when the description is by state (4.1) (multiplied by some wavefunction of the environment), we have

$$\hat{\rho}_s = \begin{pmatrix} |a|^2 & ab^* \\ a^*b & |b|^2 \end{pmatrix}, \tag{4.3}$$

while 'after decoherence', i.e. when the universe is described by the entangled wavefunction (4.2), it is of the form

$$\hat{\rho}_s = \begin{pmatrix} |a|^2 & 0 \\ 0 & |b|^2 \end{pmatrix}, \tag{4.4}$$

i.e. identical to the density matrix describing a classical mixture of the macroscopically distinct states 1 and 2 (a description which would of course follow equally from application of the hypothesis of macrorealism). At first sight, at least, observation of the phenomenon of QIMDS requires a reduced density matrix of the form (4.3) and is impossible for one of the form (4.4). This argument, with the conclusion that observation of QIMDS will be in practice totally impossible, must appear literally thousands of times in the literature of the last few decades on the quantum measurement problem, to the extent that it was at one time the almost universally accepted orthodoxy.

Before examining its possible weaknesses, let me mention briefly a related argument, namely that two 'states' of a system which are, from the standpoint of experiment, macroscopically distinct do not really correspond to single quantum states but to a whole collection of similar states, and that one usually has no experimental means of knowing which of these is the 'actual' state of any given member of the ensemble. Thus, if one schematically assigns a variable λ which labels these 'equivalent' states, one must in effect trace over the possible values of λ. Since (it is argued) the relative phase of a and b is likely to depend on λ in a random way, the effect of this is to produce a reduced density matrix of the form (4.4) rather than (4.3), so again QIMDS is precluded. Whether one regards this as an independent line of reasoning or rather as a variant[17] of the standard decoherence argument seems to me largely a matter of nomenclature; for the sake of economy of presentation I will do the latter.

Let us now try to assess the decoherence argument. Actually, the most economical tactic at this point would be to go directly to the results of the next section, namely that it is experimentally refuted! However, it is interesting to spend a moment enquiring why it was reasonable to anticipate this in advance of the actual experiments. In fact, the argument contains several major loopholes; I will briefly examine them in the abstract here, and refer back to some of them in the context of specific experiments in section 5. In the first place, it is perfectly possible to find macroscopic variables such that the coupling to the most 'obvious' environment is zero; such is the case for the centre-of-mass coordinate of an N-particle system falling freely in a uniform gravitational field, which is completely decoupled from the internal (relative) coordinates of the body (the 'diver's theorem'). Secondly, by cryogenic or other techniques it may be possible to effectively remove certain important parts of the environment

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[17] Since in practice our lack of knowledge of the relative phase is likely to be a consequence, *inter alia*, of the previous interactions of the system with its environment.

entirely. As an example, consider the normal electrons in a superconductor, which *prima facie* provide an extremely efficient mechanism of decoherence for the motion of the 'macroscopic' (flux; see section 5.4) variable: although at temperatures of the order of a degree this almost invariably destroys QIMDS completely, the density of the normal component, and hence its effect, attenuates very rapidly with decreasing temperature, and indeed it is easy to convince oneself that in a 1 cm^3 block of Nb at 5 mK there is not even one normal electron!

However, probably the most serious flaw in the standard decoherence argument is its failure to distinguish between the phenomena of 'false' and 'true' decoherence (or, what is essentially the same thing, between adiabatic and irreversible interactions with the environment). Consider, for example, the case of a U atom in a Young's-slits experiment, and suppose (actually unrealistically) that the only thing we can measure is the position of the nucleus, while the behaviour of the electrons is not directly accessible to us. The nucleus is then the 'system' and the electrons the 'environment'. If we denote the nuclear coordinate as R and that of the electrons collectively as ρ, then at the intermediate screen the wavefunction of the atomic ensemble is exactly of the form (4.2), and thus, since the electron states are orthogonal to a high degree of approximation, the reduced density matrix of the nucleus is of the 'completely decohered' form (4.4). Does this mean that (even in this hypothetical case) we would not expect to observe interference effects at the final screen? Of course not! The electrons follow the nucleus adiabatically, so by the time the two branches of $\Psi(R)$ have reconverged at the final screen, so have the two functions $\chi_{1,2}(\rho)$ describing the electronic cloud; the fringe visibility should still be 100%. On the other hand, if we imagine for example conditions near the intermediate slits such that on passing through the left slit the U atom is guaranteed to emit a (real) photon, while on passing through the right slit it is guaranteed not to, then we can say for sure that no interference pattern will be seen on the final screen—this is 'true' decoherence (the photon never as it were returns!), as opposed to the 'false' variety related to the electron cloud.

In fact, we can formulate a more generic argument concerning the observation of QIMDS. How, in practice, can we achieve this? By definition, when a superposition has a disconnectivity $\sim N$, then to distinguish it experimentally from the corresponding mixture we need to measure N-particle correlations. Now in fact physicists, at least in 2001, rarely measure even three- or four-particle correlations and almost never measure anything of higher order[18]. So how in the world can we detect a superposition with disconnectivity $\sim 10^{10}$, say? The answer is that while we do not have at our disposal an operator which is sensitive to 10^{10}-particle correlations, Nature does—it is called the time evolution operator $\hat{U}(t)$! As we shall see, virtually all relevant experiments on QIMDS to date have exploited the action of this operator, whether directly (as in Young's-slits-type experiments) or in a more subtle way.

Once one has realized this, it is rather obvious that the mere fact that the reduced density matrix of the system is of the form (4.4) at time t in no way implies that no interference can be seen at times later than t—it is entirely conceivable, and indeed quite probable, that the time evolution operator includes interactions between the system and the environment which can reverse the (false) decoherence (as it obviously does in the U-atom example cited above). It is only when an irreversible system–environment interaction (such as the emission of a real photon) takes place that 'true' decoherence occurs.

Of course, one can get only so far with qualitative, hand-waving arguments of the type used above. To justify the claim that the attempt to observe QIMDS in a specific physical system is not ridiculous, one needs to do a real QM calculation for that system which takes adequate account of the system–environment interaction. But now we face, apparently, a major

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[18] Although various theoretical papers in the quantum-optics literature have proposed to do so.

difficulty: the situation in condensed-matter physics (which is the area of most though not all relevant experiments) is totally different from that in, say, atomic physics, in that in the latter case one often has confidence that one knows the exact Hamiltonian, not just for the system in isolation, but for its interaction with its environment, e.g. the radiation field (and of course for that environment itself). By contrast, in condensed-matter systems not only is the interaction with the environment much stronger, but the fine details of the Hamiltonian are typically quite unknown. Indeed, the opinion is sometimes expressed that so great is our overall confidence in the universal validity of the formalism of QM that no experiment whatever on a condensed-matter system could ever shake it, since we will always be able to attribute any discrepancy between the QM predictions and the experimental result to an inadequate knowledge of the relevant Hamiltonian. Needless to say, were we to embrace this pessimistic point of view the whole programme outlined in this section would become rather pointless.

It turns out that there is a serendipitous way out of this difficulty[19]. Let us ask the question: rather than inferring the quantum dynamics of the system from some (unreliably guessed) microscopic Hamiltonian, can we infer it from the experimentally observed dynamics in the classical regime? (meaning the regime in which the predictions of QM are negligibly different from those of Newtonian or other classical physics). Remarkably, it has become increasingly plausible over the last 20 years that the answer is yes: what we can do is to study the experimentally observed dynamics of the system in the classical regime (where no subtle questions of interpretation arise) and derive from this, not certainly the complete structure of the microscopic Hamiltonian, but sufficient features of it to enable us to make predictions with some confidence about the behaviour with respect to essentially QM phenomena such as QIMDS. The possibility of doing this is related to the following circumstances: generally speaking, the 'environments' of the systems used in attempts to observe QIMDS (or more generally QM behaviour of a macroscopic variable) are themselves macroscopic, and under these conditions one can argue that any one environmental degree of freedom will be only weakly probed by the motion of the system. This then justifies the representation of the environment as a bath of simple harmonic oscillators (an idea which goes back at least to Feynman and Vernon [23]), with a coupling which is linear in the oscillator coordinates though for the general case if may be highly non-linear with respect to the system coordinate (i.e. the macrovariable). Now, the crucial point is that the (smoothed) spectrum of the system–oscillator coupling (usually denoted as $J(\omega)$ in the literature) is sufficient to determine (of course when supplemented by the Hamiltonian of the 'isolated' system) both the dynamics of the system in the classical regime and its characteristically quantum mechanical behaviour such as tunnelling through a classically forbidden region or QIMDS. This suggests that it might be possible to reverse, as it were, part of the argument so as to deduce the quantity $J(\omega)$ from the classical dynamics and then apply it to the quantum behaviour. Indeed it turns out, rather remarkably, that under a set of generic and not terribly restrictive conditions this is possible (24)—that is, we can use the experimentally observed dynamics in the classical regime to make unambiguous and parameter-free predictions of the quantum behaviour. In this way we avoid ever having to get involved explicitly with the microscopic details of the Hamiltonian describing the environment or its interaction with the system.

The above is, of course, theory, and one might ask whether it has been possible to put it to a meaningful experimental test. The answer is yes, at least to an extent. In fact, part of the point of 'stage 1' of the programme envisaged above is precisely to test how well the classical–quantum mapping just outlined actually works. I will discuss this further in section 5, but note here that at least in the specific case of superconducting devices, the agreement of the

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[19] For a considerably more detailed discussion of the ensuing material, see [21, 22].

experimental data on tunnelling of a macroscopic variable out of a metastable well with the parameter-free predictions of the theory is quite impressive; see in particular [25,26]. Needless to say, that agreement does not prove that the idea is completely foolproof, and we will have to return to that question in section 6.

5. The present experimental situation

Perhaps fortunately for the progress of the topic reviewed in this paper, experimental physicists tend not to be the most avid or credulous readers of the quantum measurement literature, and the scepticism expressed in much of that literature as to the possibility of ever observing QIMDS has not prevented groups in several different sub-fields of physics from trying experiments in this direction. Before addressing those experiments which give explicit evidence for QIMDS (or more generally for the quantum behaviour of a macroscopic variable), I would like to mention briefly two groups of recent experiments which, while not at the 'macroscopic' level in the sense defined in section 3, nevertheless extend the quantitative verification of the prediction of QM in this direction and have much in common conceptually with the experiments discussed later.

The first is the elegant set of experiments [27] conducted at the ENS in Paris on the entanglement of single atoms with the electromagnetic field in a microwave cavity and the resultant superposition states of that field. I will not go into the details of the procedure for producing such superpositions (which is itself a beautiful application of basic quantum mechanical principles), but the upshot is that one ends up, initially, with a superposition of the form

$$\psi = a|\alpha\rangle + b|\beta\rangle \tag{5.1}$$

where $|\alpha\rangle$ and $|\beta\rangle$ represent approximately coherent states of the electromagnetic field in the cavity[20] with complex amplitude α and β respectively; the difference $|\alpha - \beta|^2$ corresponds to a number of photons N typically in the range 5–10, so any natural definition of the disconnectivity (in terms of photons as the 'basic units') should give a value of this order. As mentioned in section 4, the cavity field is not substantially entangled with the electrons in the walls, so the 'total' D is indeed of this (microscopic) order. That the correct description is indeed initially the superposition (5.1) of the coherent states $|\alpha\rangle$ and $|\beta\rangle$ rather than a mixture of them can be verified by observing the behaviour of a second set of atoms fired through the cavity; even more pleasingly, one finds that as time goes on the superposition is gradually converted into a mixture, i.e. 'decoheres', at a rate proportional to $|\alpha - \beta|^2$ just as theory predicts[21]. While the general framework of the theory of decoherence has been widely accepted for many years, these experiments are probably the most direct and quantitative test of it to date.

A second dramatic set of experiments [28] relates to a pair of metallic grains ('boxes') each of which is superconducting. In this system the rather large capacitance energy ensures that transfer of a large number of electrons from one box to the other is energetically unfavourable, and in fact the externally applied voltages etc are engineered in such a way that only a pair of states—call them $|L\rangle$ and $|R\rangle$—which differ in the number of electrons in (say) the right box[22] by 2 are relevant at low energies. One can think of the two states $|L\rangle$ and $|R\rangle$ as

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[20] The atoms, which are essential at an intermediate stage of the process of generating states like (5.1), have by this stage been 'observed' and so are no longer entangled with the states of the field.

[21] The most obvious source of decoherence is the weak dissipation induced by the electromagnetic field in the cavity walls, and the observed rates are consistent with this hypothesis.

[22] Transfer of a single electron costs an energy of the order of the BCS energy gap ($\sim$ a few kelvins), while transfer of two electrons (a Cooper pair) does not.

corresponding to the localization of the 'last' Cooper pair (two electrons) in the left and the right box respectively. If the two boxes are indefinitely far apart, these states are eigenstates of the many-body Hamiltonian and can be made exactly degenerate. However, in the presence of a non-zero degree of contact between the boxes (e.g. through an insulating barrier), the Josephson tunnelling can transfer a Cooper pair, i.e. convert $|L\rangle$ into $|R\rangle$ and vice versa. Thus, to a good approximation, this complicated many-body system can be regarded effectively as a simple two-state system, with a 'tunnel splitting' given by the Josephson matrix element Δ (not to be confused with the single-particle BCS energy gap). Then, if the bias is adjusted such that in the absence of tunnelling $|L\rangle$ and $|R\rangle$ are degenerate, and one starts the system in (say) the state $|L\rangle$, then the subsequent wavefunction is time dependent:

$$\psi(t) \sim \cos(\Delta t/2)|L\rangle + \sin(\Delta t/2)|R\rangle \tag{5.2}$$

so the probability $P_L(t)$ of finding the extra Cooper pair on (say) the left is oscillatory:

$$P_L(t) = \tfrac{1}{2}(1 + \cos \Delta t). \tag{5.3}$$

This probability can be measured by adding a weak mechanism for a particle in (say) the left box, but not in the right box, to tunnel out into an external circuit, and when the histogram of the resultant current is plotted, a beautiful oscillatory pattern corresponding to (5.3) is seen, with up to 20 periods observable.

Two points are particularly worth noting about this experiment. First, one might perhaps be sceptical that a coherent oscillation can persist even in the presence of a 'measurement' of which state, $|L\rangle$ or $|R\rangle$, the pair is occupying; have we not all been told that any attempt to 'measure' which of the two states of a superposition is realized automatically destroys the superposition? The resolution [28] is that the measurement is a very 'weak' one, which has an efficiency very much less than 100%, and so does not interfere appreciably with the coherent tunnelling process, while at the same time the results for a sufficiently large (time) ensemble are statistically significant. Secondly, while the value of the 'extensive difference' realized in this experiment is associated with the electric dipole moment and of the order of 10^5, with the result that it may reasonably be characterized as 'macroscopic', the disconnectivity D is only 2, or very close to it. This might at first sight seem surprising, since it is tempting to argue that even though it is only a single Cooper pair which tunnels, all the $\sim 10^9$ electrons within the box in which the pair arrives must recoil, and thus their state should be entangled with that of the pair; however, just as in the microwave-cavity case discussed in section 4, an explicit calculation shows that the degree of orthogonality of the two relevant states of this 'environment' is very small.

I now turn to experiments in which the states involved are by some reasonable criterion 'macroscopically' distinct. Such experiments have been conducted on a variety of systems[23], but in some cases are currently still at what I called in section 4 'stage 1' of the programme. I will discuss here only those cases in which 'stage 2' has been reached, that is, in which more or less direct evidence has been claimed for the phenomenon of QIMDS. To the best of my knowledge, such a stage has been reached to date in four types of system: large molecules in free space, magnetic biomolecules, quantum-optical systems and superconducting devices. I will discuss these in turn.

5.1. Diffraction of complex molecules in free space

Experiments on the diffraction of objects more complex than single atoms (actually H_2 molecules) are almost as old as QM itself, but until about two years ago the most sophisticated

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[23] And proposed on yet others.

systems for which such diffraction had been observed were loosely bound complexes of up to about eight ^{4}He atoms or H_2 molecules (corresponding, in the notation of section 3, to a disconnectivity of about 30). In 1999 a spectacular advance in this field was achieved by Arndt *et al* [29], who succeeded in observing diffraction effects with a beam of C_{60} (and subsequently also C_{70}) molecules. While the details of the preparation and detection techniques used (for which the reader is referred to the original paper and to [30]) are highly non-trivial, conceptually the experiment is a simple generalization of the classic Young's-slits experiment, with slits spaced by 100 nm; despite the rather large velocity spread in the incoming beam (cf below), the diffracted intensity as a function of angle clearly shows a central peak flanked by two first-order satellites (see figure 2 of [29]). The data are well fitted within the error bars by a standard quantum mechanical calculation with two fitting parameters. In terms of the measures defined in section 3, the extensive difference Λ may be estimated at around 10^6, while the disconnectivity D is by the usual 'count' 1080 for C_{60} (and slightly larger for C_{70}).

Two features of this experiment are in particular remarkable. In the first place, in strong contrast to the experiments described in subsections 2 and 4 of this section, which require cryogenic techniques, the ensemble (molecular beam) is prepared in an oven at 900–1000 K and no monochromator is used, and the velocities therefore have a considerable spread. That this spread (which is a special case of the 'uncertainty in initial conditions' discussed in section 4) does not completely wash out the (satellite) peaks and troughs in the diffraction pattern is associated with the fact that the Maxwell–Boltzmann exponential factor is peaked not around zero but around a finite effective velocity v_0, i.e. the (independently measured) velocity distribution in the incident beam is of the form

$$f(v) = Av^3 \exp{-(v - v_0)^2/v_m^2} \qquad (5.4)$$

with $v_0 \sim 1.8v_m$.

Secondly, the molecules of the beam are by no means in their ground state with respect to their internal degrees of freedom; indeed, it is estimated that the average energy associated with these is approximately 5.8 eV, distributed over the 174 degrees of freedom of vibration of the C_{60} molecule. Moreover, four of these modes are infrared active, i.e. couple strongly to the black-body radiation field, and from the known Einstein coefficients it is estimated that three or four quanta are emitted (and absorbed) during the passage of a molecule through the apparatus. Thus, the interaction of the system with its 'environment' is by no means 'weak'!

Why, nevertheless, does decoherence not destroy all possibility of observing QIMDS in this experiment? The main reason, of course, is that in free space[24] the motion of the centre of mass is not directly coupled to the internal degrees of freedom (the 'diver's theorem'; cf section 4). However, this observation does not in itself eliminate the problem, since there is an indirect coupling via the interaction of the infrared-active vibrational modes with the black-body radiation field; if the final state of the radiation field is appreciably different depending on which slit the molecule passed through, this should be enough to decohere the superposition and thus destroy the diffraction pattern. As emphasized by the authors of the experiment [29], what saves the day is the fact that the wavelength of the photons emitted in the relevant transitions is much larger than the inter-slit distance, so the final state of the radiation field is nearly independent of which slit was traversed. (Compare the classic 'γ-ray microscope' thought-experiment [31].)

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[24] Close to the slit edges there should be some coupling; presumably it is not strong enough to produce appreciable decoherence.

5.2. Magnetic biomolecules

Until the experiments described in the last subsection, the most plausible candidate for the claim of observation of QIMDS was probably the system formed by the magnetic moments in ferritin molecules. The ferritin molecule, which is naturally produced in biological systems and in particular in the horse spleen, consists of a protein sheath ('apoferritin') surrounding a cavity of dimension $\sim$7.5 nm, which in the naturally occurring species is filled with an iron compound ($9Fe_2O_3 \cdot 9H_2O$) containing about 4500 Fe^{3+} ions; by suitable chemical techniques it is possible to produce either pure apoferritin (with the cavity vacant) or variants in which the number of Fe ions can be as low as 100. Despite the complicated nature of this system, it has been the subject of many investigations of its structure and magnetic properties, and the general belief is that in natural ferritin the core of Fe^{3+} ions is predominantly ordered antiferromagnetically, with however a few of the spins on the surface providing a residual magnetic moment which is estimated at approximately 220 μ_B. Because of the crystalline anisotropy, the ground state of an isolated core is not expected to be invariant under rotation; however, in view of the invariance under time reversal, a state with total spin $S = |S|\hat{n}$ lying along the easy axis should be degenerate with that corresponding to $S = -|S|\hat{n}$ in the absence of tunnelling, and it is these two states whose superposition is sought. In the real-life situation the situation is more complicated, since the magnetization of neighbouring cores will be coupled by the dipole force; however, this effect can be substantially reduced by diluting the naturally occurring compound with pure apoferritin.

In order to draw any conclusions about the possible superposition and interference of the two states of an individual ferritin core corresponding to $S = \pm|S|\hat{n}$, one needs of course a theory of the dynamics of the collective spin which will include possible effects of quantum tunnelling; crudely speaking, since the magnitude $|S|$ of the total spin S is constant to a good approximation, to make a transition from $\hat{n}$ to $-\hat{n}$ the spin must tunnel through regions where, because of the crystalline anisotropy, it feels an energy barrier. Because of the rather unusual nature of the spin variable from the point of view of quantum mechanics, the theory of such tunnelling processes is not a trivial generalization of that applied to other collective variables, such as the flux in SQUIDs (see the next subsection), but by the early 1990s it had been well developed and applied to explain various phenomena connected in the magnetic relaxation at low temperatures; see e.g. [32]. (In the language of section 4, this is 'stage 1' of the programme.) However, in view (*inter alia*) of the rather complicated magnetic structures of many of the systems involved, quantitative tests of the predictions of theory against experiment at the reasonably 'macroscopic' level[25] have not to date proved easy, and one's confidence in the predictions has to rest largely on *a priori* considerations.

An important conclusion of the theory of magnetic quantum tunnelling is that for a purely ferromagnetic system tunnelling requires biaxial anisotropy[26], and the WKB exponent is $N(K/K_\perp)^{1/2}$, where $K_\perp$ is the coefficient of this anisotropy, K that of the uniaxial term and N the total number of spins. By contrast, in a purely antiferromagnetic system tunnelling between two 'easy' configurations related by time reversal requires only uniaxial anisotropy, and the formula for the splitting of the ground state by such tunnelling is (in the absence of considerations related to Kramers' theorem[27]) given by

$$\Delta \sim \hbar\omega_0 \exp(-N\sqrt{K/J}) \qquad (5.5)$$

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[25] However, at the 'mesoscopic' level of molecules such as Mn_{12} acetate and Fe_8, quite impressive quantitative agreement has been obtained; see e.g. [33].

[26] Since with uniaxial anisotropy and the easy axis along $\hat{z}$, S_z is conserved.

[27] In fact, there are some very interesting and still not wholly understood issues related to Kramers' theorem in this context; see in particular [34] and [35].

where N is the total number of spins involved, K the (uniaxial) anisotropy energy per spin, J the (isotropic) exchange energy and ω_0 an 'attempt' frequency of order $\sqrt{KJ}/\hbar$. Because J tends to be substantially larger than the biaxial anisotropy $K_\perp$, the tunnelling in AF systems is generally considerably larger than in ferromagnetic ones. In the case of ferritin, which as described above is primarily antiferromagnetic but with a small residual total moment, the data have been analysed under the assumption that the splitting between the two 'ground states' $|+\rangle$ and $|-\rangle$ related by time reversal is given, at least to a first approximation, by the AF formula (5.5), with the residual moment acting as a 'tag' of the state (and, in the presence of an external magnetic field H, providing of course an additional coupling energy—$S \cdot H$ which will split the two ground states even in the absence of tunnelling.) Arguments which support this analysis are given in [36].

In their original experiments [27] on an ensemble of (biological) ferritin particles diluted with apoferritin, Awschalom *et al* measured the magnetic susceptibility and associated noise spectrum in the kilohertz frequency range. Above about 200 mK, the data are featureless, but below this temperature a fairly sharp peak appears in both measurements at a frequency of about 940 kHz. (This feature is not seen for less highly diluted samples.) They attribute this peak to coherent collective tunnelling ('MQC') of the spins between the two 'ground states', an interpretation which, if correct, would be fairly direct evidence for QIMDS, since to produce such an oscillation the state at intermediate times would have to be a superposition of the form $\alpha(t)|+\rangle + \beta(t)|-\rangle$, and argue that existing estimates of the parameters are consistent with the splitting (5.5) indeed being of the order of the observed frequency. Since each of the Fe^{3+} spins is reversed in the transition $\hat{n} \to -\hat{n}$, the disconnectivity of the inferred superposition would be ~ 4500; if we are prepared to regard the staggered magnetization as an 'extensive' variable for the purposes of the definition of the 'extensive difference' Λ, then the latter would also be ~ 4500; otherwise it is ~ 200.

Needless to say, the mere observation of a resonance of the total magnetization with a frequency of the order of that predicted by equation (5.5) would not by itself be particularly convincing evidence for MQC. However, there are two features of these and subsequent experiments on diluted ferritin which support the above interpretation, at least qualitatively. The first is the dependence on the external field H. If we assume for the moment (unrealistically; cf below) that the easy axis $\hat{n}$ is parallel to H for all of the ferritin molecules, then a simple two-state analysis predicts that if the above interpretation is correct, the field dependence of the resonance frequency ω_m would have the Pythagorean form

$$\omega_m = (\omega_0^2 + M^2 H^2)^{1/2}$$

where ω_0 is the zero-field value 940 kHz. Although the data do not fit this prediction perfectly, the qualitative trend is similar (see e.g. figure 2(*a*) of [37]), and in particular the value of H at which substantial deviation from the zero-field value begins is indeed of order 10^{-4} G, which is consistent with ω_0/M if $M \approx 217 \, \mu_B$. This feature of the data suggests rather strongly that whatever the detailed nature of the resonance, the relevant variable is indeed the total spin S of the core rather than (say) that of an individual Fe^{3+} ion.

The second feature which lends support to the identification of the observed resonance with that associated with MQC is the dependence, investigated in a subsequent experiment [39], of the frequency on the number N of Fe^{3+} spins in the apoferritin cavity, a quantity which as already indicated can be varied by the experimenter. According to formula (5.5), one would expect the relation

$$\ln \omega_{res} = a - bN$$

and this seems at least crudely consistent with the data[28] (see figure 3 of [38], where ω_{rs} varies by more than two orders of magnitude). Again, it seems rather difficult to think of an explanation of this feature which does not involve the collective quantum tunnelling of $\sim N$ spins.

Despite the above considerations, the interpretation given by Awschalom *et al* of their raw data in terms of MQC has been subjected to severe criticism: see [39] and [40]. The principal grounds for the criticism of [39] are:

(1) that the data do not satisfy the Pythagorean law (above) quantitatively;
(2) that the absorption falls off with increasing field much more slowly than predicted theoretically under the MQC hypothesis;
(3) that since in practice $\hat{n}$ is oriented in a random direction and thus $|M \cdot H|$ varies between zero and MH, the spread in frequency for $MH \gtrsim \omega_0$ should be much larger than observed;
(4) that the theoretical power absorption is only a small fraction of that observed experimentally.

Reference [40] further concludes that direct measurements of the relevant parameters leads to a tunnelling frequency more than ten orders of magnitude smaller than that observed. While the original authors have replied [41] to the criticisms of [39], their ultimate argument in favour of their original interpretation is simply that it is very difficult to think of an alternative scenario. In these circumstances one's attitude to these experiments will probably be a function of one's general philosophy of science: in the present author's case it is to reserve judgment.

5.3. Quantum-optical systems

As already noted, despite the frequent use of the words 'Schrödinger's cat' in papers in the quantum-optical literature, the actual experiments conducted in this area up to now have almost never realized values of the parameters Λ and D which by any reasonable definition are much greater than ~ 10. However, very recently a group at Aarhus has conducted an experiment [42] which, while not primarily motivated by the programme reviewed here, certainly qualifies for inclusion in this section.

The principle underlying the Aarhus experiment is the following: consider two spin-polarized atomic samples such that $J_{x1} = -J_{x2} \equiv J_x$, where J_x is assumed to be of the order of the total number N of atoms in each sample, and thus can be treated to a good approximation as a classical variable. Then, using the standard commutation relation $[J_{yj}, J_{zj}] = iJ_{xj}(j = 1, 2)$, we see that the variances $\delta J_{xj}^2 \equiv (J_{xj} - \langle J_{xj} \rangle)^2$ etc satisfy the Heisenberg-type inequality

$$\delta J_{zj}^2 + \delta J_{yj}^2 \geqslant J_{xj} \qquad (j = 1, 2) \tag{5.6}$$

(where the equality is attained for a coherent spin state). On the other hand, the y- and z-components of the total spin $J_1 = J_2$ commute, so the sum of the variances can in principle vanish; more generally, there is nothing to prevent satisfaction of the inequality

$$\delta J_z^2 + \delta J_j^2 < 2J_x \tag{5.7}$$

which when combined with (5.6) says intuitively that the variances of the quantities which refer to the total system are less than the sum of those characterizing the two subsystems. (As discussed in [42], this situation can be put in one-to-one correspondence with the one discussed in the original paper of Einstein *et al* [43] on the non-local aspects of quantum mechanics.)

Let us consider this situation from the point of view of the search for QIMDS, assuming that in a suitable experiment the inequality (5.7) is found to be satisfied. Of course, this fact alone does not demonstrate the existence of QIMDS (and would not even if the inequality (5.6)

[28] Though a sceptic might observe that were the last data point (at $V/V_0 \sim 0.2$) removed, the fit would look distinctly problematic!

was simultaneously verified experimentally); it is clear that a classical statistical model in which the yz-components of $\boldsymbol{J}_1$ and $\boldsymbol{J}_2$ are exactly anticorrelated but the direction of $\boldsymbol{J}_1$ in the yz-plane is random could in principle satisfy the two relations simultaneously. However, once we have postulated that the description of the whole system is to be in quantum mechanical terms, we can draw a non-trivial conclusion. Let us consider for simplicity the special case (not realized in the Aarhus experiment) where the LHS of (5.7) is found to be zero. Then the inequality (5.6), assuming it to be experimentally verified (or postulated *a priori*) shows that the ensemble must possess a spread of values of J_{x1} which is at least of order $J_x^{1/2}$ (and hence $\sim N^{1/2}$). On the other hand, for the LHS of (5.7) to be zero, the value of J_{x1} must be perfectly anticorrelated with that of J_{x2}. Thus the density matrix of the total system must correspond to either a superposition or a mixture of states corresponding to values of the pair of quantities (J_{x1}, J_{x2}) which differ by amounts of order $N^{1/2}$. However, it can be shown [44] that given the standard QM commutation relations of the components of $\boldsymbol{J}$, no mixture of such states could in fact satisfy (5.7). Thus, the density matrix indeed corresponds to a superposition, and thus we have realized 'stage 2' of the programme outlined in section 4, with values of the quantities Λ and D which by any reasonable definition are of order $N^{1/2}$. Clearly qualitatively similar considerations apply whenever the inequality (5.7) is violated by an amount of order 1, even if the LHS is not zero.

In the Aarhus experiment, each subsystem consisted of around 10^{12} Cs atoms. A state of the kind discussed above was prepared by an 'entangling' optical pulse, and the satisfaction of the inequality (5.7) was demonstrated by a second ('verifying') pulse (see [42] for details). The measured value of the ratio of the LHS and RHS of (5.7) was about 65%, corresponding to a 'degree of entanglement' (see [42]) of about 35%. This entanglement was found to persist for a time of order 0.5 ms following the 'entangling' pulse.

Thus, in this experiment it seems that stage 2 of the programme of section 4 has been realized, with values of Λ and D which are of order 10^6. Because of the rather 'clean' nature of these quantum-optical systems, it is possible that they may in future become competitive with SQUIDs (see the next subsection) as the experimental systems of choice in the search for QIMDS.

5.4. Superconducting devices

Of the various areas of physics in which the search for evidence of quantum interference of macroscopically distinct states has been pursued, that of superconducting devices has been the one in which the programme has been both most ambitious and most systematic. That these devices, in all cases based on the Josephson effect, are promising candidates for the observation of QIMDS has been clear for over 20 years now. Their advantages include the fact that the classical dynamics of the relevant macroscopic variable (the trapped flux; see below) is believed to be excellently understood; that the 'intrinsic' dissipation associated with the motion of this variable can be made extremely weak (by condensed-matter standards, at least!) by going to temperatures which, while low by everyday standards, are nowadays routinely attainable; and that this variable can be addressed by electromagnetic means using technology already familiar to electrical engineers. However, possibly the most compelling advantage of these systems relative to those reviewed in subsections 1 and 2 has to do with the way in which the difficulty of observation of QIMDS scales with the 'measures of macroscopicness' Λ and D defined in section 3. In the case of free-space molecular diffraction, it is clear that the angular fringe spacing is inversely proportional to the mass of the molecules involved (hence, roughly speaking, to the disconnectivity D). Similarly, in the case of magnetic tunnelling of the type discussed in subsection 2, equation (5.5) shows immediately that the tunnelling splitting

falls off exponentially with the total number of particles N ($\approx D$) which tunnel collectively, and in the quantum-optical case of subsection 3 the value of D increases only as $N^{1/2}$. In the superconducting-device case the situation is rather different and more encouraging: quite generically, what is remarkable about the Josephson effect is that through its use the motion of a macroscopic variable (such as the flux in a ring visible to the naked eye) can be controlled by an energy which is itself of the order of the thermal energy of a single atom at room temperature, i.e. by any reasonable definition microscopic. This energy is not associated with the bulk ring itself, but rather with the Josephson junction, and thus is independent of the size (circumference) of the ring. Certainly, the matrix element for quantum tunnelling between the macroscopically distinct states of the bulk ring has the standard WKB form with an exponent which is proportional to the square root of an effective 'mass' (see equation (5.20) below), but, at least in the regime accessed by the experiments to be described, this 'mass' is again a characteristic of the junction and so does not scale with the ring size. Thus it is not surprising that the values of the parameters Λ and D which have been achieved with superconducting devices exceed by several orders of magnitude those currently attained in free-space molecular diffraction, in magnetic biomolecules or even in quantum-optical systems, and I make no apology for devoting more space to them.

I now briefly outline the description of Josephson devices in terms of a macroscopic quantum variable; for a considerably more detailed account, see [22]. According to the standard theory of superconductivity (see e.g. [45]), this state is characterized by the formation of Cooper pairs, which we may think of for present purposes as a sort of di-electronic molecules, which are automatically 'Bose-condensed' into a single two-particle state whose wavefunction[29] we denote as $\Psi(r_1, r_2)$. Any dependence of Ψ on the relative coordinate $r_1 - r_2$ is irrelevant in the present context, so I shall treat Ψ as a function only of the centre-of-mass coordinate of the 'molecule', $R \equiv \frac{1}{2}(r_1 + r_2)$:

$$\Psi = \Psi(R) \equiv |\Psi(R)| \exp \mathrm{i}\varphi(R) \tag{5.8}$$

where it is the phase $\varphi(R)$ which will play the essential role below. In addition to the electrons which are bound into Cooper pairs and constitute the 'superfluid (superconducting)' component of the system, there will in general be some unpaired electrons which constitute the 'normal' component. However, an energy gap Δ_{BCS}, of the order of the transition temperature, T_c, separates the energy of an unpaired electron from that of one bound in a Cooper pair, and as a result the fraction of electrons which are 'normal' is proportional to $\exp(-\Delta_{BCS}/T)$; as already noted, for Nb ($T_c \sim 9$ K) at say 5 mK, this fraction is so small that even a macroscopic (say ~ 1 cm^3) block of the metal contains not a single normal electron! Thus I shall from now on ignore any effects of the normal electrons in the bulk superconductor.

I will use for illustration the simplest form of Josephson device which displays all the features of interest in the present context, namely a single rf (or single-junction) SQUID ring of the type illustrated in figure 1. This is simply a bulk superconducting ring (typically of thickness much larger than the London penetration depth (on which, see below)) interrupted by a single Josephson junction; for simplicity I will assume that the latter is constituted by a thin (typically $\sim$30–50 Å) layer of oxide (the 'tunnel–oxide' junction), which allows electrons to pass through by tunnelling. According to the standard theory of Josephson tunnelling (see e.g. [47]), any discontinuity $\Delta\varphi$ of the phase $\varphi(R)$ of the Cooper pairs across the junction is associated with an energy of the form

$$E_J = -\frac{I_c\varphi_0}{2\pi} \cos \Delta\varphi \tag{5.9}$$

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[29] In practice one deals with the so-called 'anomalous average' which, while not quite a wavefunction in the literal sense, plays the role of one; see e.g. [46], section 10.4.
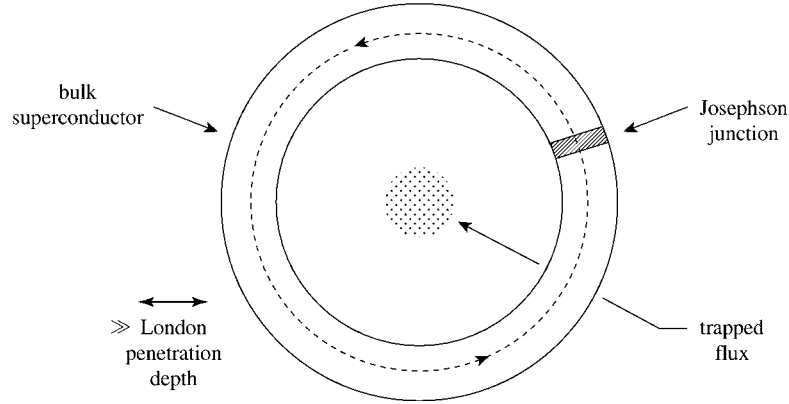
Figure 1. A single rf (or single-junction) SQUID ring.

where $\varphi_0 \equiv h/2e$ is the superconducting flux quantum, and I_c has the physical significance of the critical current—that is, the maximum current which can be transmitted by the junction without dissipation. (However, in the present context we can just regard I_c as a convenient parametrization of the locking energy E_J.) In the experiments to be described, I_c is of the order of 1 μA, so the locking (Josephson) energy E_J is a few tens of kelvins.

Like any quantum phase, the quantity $\varphi(R)$ must be single valued modulo 2π, and therefore any drop $\Delta\varphi$ across the junction must be compensated by a corresponding variation around the bulk ring. To calculate the latter, it is convenient to choose a path which lies much more than a penetration depth from the surface. Along such a path the electric current must be zero (see e.g. [47]) and since quite generally the current carried by the pairs is proportional to $\nabla\varphi - (2e/\hbar)A(r)$ where $A(r)$ is the vector potential, it follows that $\nabla\varphi = 2eA(r)/\hbar$ and hence that the total drop in φ around the bulk ring is $2\pi\Phi/\varphi_0$, when Φ is the total flux trapped through the ring (we neglect the very small contribution to Φ from the barrier region itself). Since as remarked the phase must be well defined modulo 2π, we get a fundamental relation between the flux Φ and the phase drop $\Delta\varphi$ across the junction:

$$\Delta\varphi + 2\pi\Phi/\varphi_0 = 2n\pi \tag{5.10}$$

and thus the behaviour of the Cooper pairs is characterized uniquely by the single variable Φ, which will serve as the 'macroscopic variable' for our considerations below.

It is important to appreciate that although the whole basis of our considerations is quantum mechanical, at the present stage of the argument we are still treating the quantity Φ as a classical variable which parametrizes the Cooper-pair wavefunction: in fact, schematically, the latter is (neglecting normalization, antisymmetrization, the spin degree of freedom and other technicalities)

$$\Psi(r_1 r_2 \cdots r_N, t) \sim \chi(r_1 r_2 : \Phi(t))\chi(r_3 r_4 : \Phi(t)) \cdots \chi(r_{n-1} r_n : \Phi(t)) \equiv \Psi_\Phi \tag{5.11}$$

where if we take a path such as that shown in figure 1 and denote the angular coordinate corresponding to the COM variable $r \equiv \frac{1}{2}(r_1 + r_2)$ by θ, we have (again schematically)

$$\chi(r_1 r_2 : \Phi(t)) \sim \exp i(\Phi(t)/\varphi_0)\theta. \tag{5.12}$$

To obtain the total (relevant) potential energy of the ring as a function of Φ, we substitute (5.9) into (5.10) and add the purely electromagnetic term generated by the finite self-inductance L of the ring, namely $\frac{1}{2}LI^2 = (\Phi - \Phi_{ext})^2/2L$ where Φ_{ext} is any externally applied flux. Thus we obtain the formula

$$U(\Phi) = (\Phi - \Phi_{ext})^2/2L - (I_c\varphi_0/2\pi)\cos(2\pi\Phi/\varphi_0). \tag{5.13}$$

It may be verified that for $\beta_L \equiv 2\pi L I_c/\varphi_0 > 1$ and suitable values of Φ_{ext}, expression (5.13) has more than one local minimum, which therefore can correspond to metastable configurations of the Cooper pairs.

In addition to the 'potential' energy as given by expression (5.13), we might expect intuitively that there might be a 'kinetic' energy associated with any change of Φ in time. This is indeed so, for purely electromagnetic reasons: a time variation of the flux Φ through the ring corresponds to generation of an emf around it, which in turn leads to a build-up of charge and the associated electrostatic energy. Although from a microscopic point of view this energy is 'potential', when expressed in terms of Φ it comes out in the form $\frac{1}{2}C\dot{\Phi}^2$, and it is therefore natural to regard it as 'kinetic' and the capacitance C as the effective 'mass' associated with Φ. The question of the actual value of C is rather delicate in the general case (see [22], section (3.4)); however, provided that the effective capacitance of the junction itself is much larger than the geometrical capacitance of the ring (which is true for the experiments discussed below), it is safe to take C as the former. With that choice, the 'momentum' p_Φ conjugate to Φ, $C\dot{\Phi}$, turns out to be simply the charge imbalance developed across the junction. The complete Hamiltonian is thus

$$H = \frac{p_\Phi^2}{2C} + \left\{ \frac{(\Phi - \Phi_{ext}^{(t)})^2}{2L} - \frac{I_c \varphi_0}{2\pi} \cos(2\pi \Phi/\varphi_0) \right\}, \qquad p_\Phi \equiv C\dot{\Phi}. \quad (5.14)$$

The Hamiltonian (5.14) is of course conservative: to incorporate the effects of dissipation one usually adds to the Newtonian (classical) equation of motion a phenomenological term of the form $\dot{\Phi}/R$, where R is the effective resistance shunting the junction, so that it reads

$$C\ddot{\Phi} + \dot{\Phi}/R + \partial U/\partial \Phi = 0. \tag{5.15}$$

This purely classical equation of motion[30] for the flux has been very widely used in the description of practical SQUIDs, and appears to be very well confirmed by experiment; see e.g. [47]. Thus we have a firm classical jumping-off point for our discussion of macroscopic quantum superposition.

Conceptually, the procedure necessary to go over from a classical treatment of the flux to a quantum one is straightforward: rather than assuming that the many-body wavefunction is of the simple form (5.11), i.e. corresponds at any given time to a single unique value of Φ, we allow ourselves to consider superpositions of states with different values of Φ: schematically,

$$\Psi(r_1 r_2 \cdots r_N : t) = \int d\Phi \, C(\Phi : t)\Psi_\Phi(r_1 r_2 \cdots r_N) \tag{5.16}$$

where the complex coefficient $C(\Phi : t)$ is simply the analogue of the 'wavefunction' $\psi(x, t)$ in a single-particle Schrödinger problem, and where we have put the time dependence into the coefficient C in accordance with the normal practice in the Schrödinger representation. Equation (5.11) is the special case of (5.16) in which $C(\Phi : t)$ is a δ-function. It is clear intuitively that forms of (5.16) in which $C(\Phi : t)$ is substantial for two or more different regions of Φ which are different by an amount $\sim \varphi_0$ will correspond to a large value of the disconnectivity D.

To the extent that the simple conservative Hamiltonian (5.14) is an adequate description, the transition from classical to quantum dynamics is attained simply by replacing H by $i\hbar \, \partial/\partial t$ and the classical quantity p_Φ^2 by the differential operator $-\hbar^2 \, \partial^2/\partial\Phi^2$, and allowing (5.14) to operate on the set of complex amplitudes ('wavefunction') $C(\Phi : t)$; in fact, replacing the

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[30] The model embodied in equation (5.14) is usually called the RSJ(C) model ('resistively shunted junction with capacitance'). Of course, in some cases it may be necessary to generalize so as to allow (for example) the shunting resistance R to be frequency dependent, or for the external circuitry.

latter by the more natural notation $\psi(\Phi : t)$ (since there is by now, we hope, no danger of confusion in this notation), we have

$$i\hbar \frac{\partial \psi(\Phi : t)}{\partial t} = \left\{ -\frac{\hbar^2}{2C} \frac{\partial^2}{\partial \Phi^2} + U(\Phi : t) \right\} \psi(\Phi : t). \tag{5.17}$$

We could then proceed to discuss phenomena such as level quantization, tunnelling through a classically forbidden barrier and so on.

In real life, of course, such a procedure is much too naive. In the classical case we know that the dissipative term in (5.15) is often very important and can even change the behaviour qualitatively; what are the corresponding effects in quantum mechanics? It was precisely to answer this question that the method described in section 4, which exploits a serendipitous classical–quantum correspondence, was developed. This is a whole topic in itself, and I refer the reader for example to [24] for an extended discussion; here it suffices to state the conclusion of the theoretical considerations, namely that under certain rather generic and plausible assumptions a complete knowledge of the classical equations of motion of the flux is sufficient for a parameter-free prediction of the behaviour in the quantum regime. If this conclusion is correct, it is significant not only because it allows the experimenter to measure, in purely classical experiments, all the parameters relevant to a projected experiment in the 'deep quantum' regime, but also because if this is done and one then were to obtain results very different from the quantum predictions, it would be impossible (or at least very much less plausible) to attribute this to the unknown effects of dissipation and/or decoherence.

However, in the early days of the subject it was not even obvious (at least to a sizable fraction of the community) that the very idea of quantizing the dynamics of a collective variable such as the flux by the simple prescription (5.17) was legitimate, let alone that the 'classical–quantum correspondence method' was viable. Thus, in the language of section 4, stage 1 of the programme lasted in the case of Josephson devices for 20 years; during this time a variety of painstaking experiments were conducted on aspects of the behaviour of the flux variable (or a related one[31]) such as tunnelling out of a metastable well, level quantization, resonant activation and incoherent relaxation of a 'two-state' configuration, and the results compared with the predictions of quantum mechanics as embodied in the above prescriptions. While there are a few aspects of the experiments which are not totally understood, I think it is fair to say that the general pattern of agreement is excellent; particularly striking in this respect are the parameter-free fits made to the data in [25] and [26], which both relate to situations where the effect of dissipation is highly non-negligible. While none of these experiments are yet at the level of 'stage 2' (provision of direct evidence for QIMDS), their results give us considerable confidence in moving on to this stage.

Within the last 18 months, in fact, two groups have provided strong and fairly direct evidence for the phenomenon of QIMDS. To understand the nature of the evidence, it may be helpful to consider initially a model experiment which is actually slightly different from either of the actual ones. Let us consider a system with $\beta_L \equiv 2\pi L I_c / \varphi_0 > 1$, and suppose that the external flux (which for the moment is treated as a c-number) is exactly half a flux quantum: $\Phi_{ext} = \frac{1}{2}\varphi_0$. Then the potential energy $U(\Phi)$ has the symmetric form shown in figure 2, with two degenerate wells, which we will label $|L\rangle$ and $|R\rangle$, separated by a classically impenetrable barrier, and the 'ground state' within each well separately will be localized as indicated in figure 2 on a scale very much smaller than the inter-well distance. Thus, $|L\rangle$ and $|R\rangle$ can form the 'macroscopically distinct' states required for our purpose. We seek evidence of the existence of superpositions of the form

$$\psi = \alpha(t)|L\rangle + \beta(t)|R\rangle \tag{5.18}$$

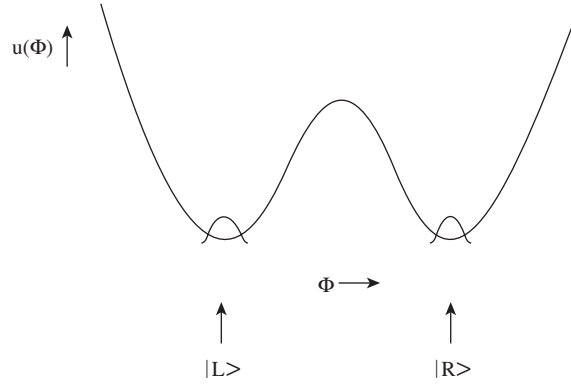[31] The phase variable in an externally biased Josephson junction.

Figure 2. The form of the potential energy $U(\Phi)$ (equation (5.13)) for external flux $\Phi_0/2$.

as distinct from classical mixtures of $|L\rangle$ and $|R\rangle$. Generally speaking, detection of such superpositions relies on the existence of a non-zero matrix element $\Delta/2$ for transitions between $|L\rangle$ and $|R\rangle$, such that in the unbiassed case considered, the effective two-state Hamiltonian takes the following form in the $|L\rangle|R\rangle$ basis (the factor of $1/2$ is inserted for convenience):

$$H_{eff} = \begin{pmatrix} 0 & \Delta/2 \\ \Delta/2 & 0 \end{pmatrix}. \tag{5.19}$$

In the physical case considered, the quantity Δ is the amplitude for tunnelling through the barrier (forbidden region of flux) and is schematically of the form

$$\Delta \sim \text{constant} \times \exp\left(-\int (2CU(\Phi))^{1/2}\,d\Phi/\hbar\right) \tag{5.20}$$

where both C and $U(\Phi)$ are primarily associated with the (physical) oxide barrier rather than the bulk ring; thus a relatively substantial value of Δ is not in itself incompatible with a fairly 'macroscopic' geometry of the circuit as a whole.

The most obvious way of searching for the effects of superpositions such as (5.18) would probably be a 'real-time' experiment in which one starts the system off in, say, $|L\rangle$, switches one's measuring apparatus off, then switches it on again at time t and detects whether it is in $|L\rangle$ or $|R\rangle$, and by making repeated runs of this type with varying values of t, plots a histogram of the probability $P_L(t)$ that the system is in $|L\rangle$ at time t. A standard quantum mechanical calculation will find that when unobserved the system is described by a wavefunction of the form (5.18), with $\alpha(t) = \cos\Delta t/2$ and $\beta(t) = \mathrm{i}\sin\Delta t/2$ (i.e. equation (5.2)) and (of course) that repeated observation at time t (with no intermediate observation) will yield $P_L(t) = |\alpha(t)|^2 = \frac{1}{2}(1 + \cos\Delta t)$ (cf equation (5.3)); thus, in particular, at time $t = \pi/\Delta$ the system will be found in the $|R\rangle$ well. Imagine on the other hand that at the intermediate time $t = \pi/(2\Delta)$ the system were not in the superposition (5.2) but in the corresponding mixture of $|L\rangle$ and $|R\rangle$ (so that the probability for each is 50%); if we take this as our initial condition and integrate the time-dependent Schrödinger equation, we find that at all subsequent times, and in particular at $t = \pi/\Delta$, the system is still in an equal-probability mixture. Thus, when interpreted according to standard QM, the fact that $P_L(t = \pi/\Delta) = 0$ is strong evidence that the state of the system at time $\pi/(2\Delta)$ was an (equal-weight) superposition of $|L\rangle$ and $|R\rangle$ rather than a mixture of these two states.

While, as described above, a 'real-time' experiment of the above nature has been successfully performed [28] for the more microscopic case of a Cooper-pair tunnelling between
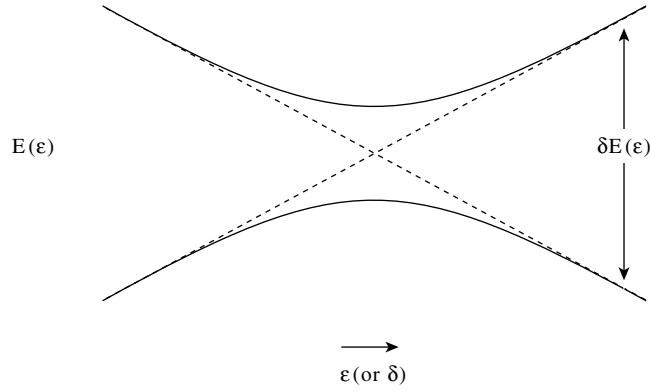
$E(\varepsilon)$ $\delta E(\varepsilon)$

ε (or δ)

Figure 3. The 'avoided-crossing' form (solid curve) and the diagonal elements of H_{eff}, i.e. $\pm\epsilon/2$ (dashed curves).

two superconducting grains (where the tunnelling is of course in real space rather than in flux space; see above), it had not until very recently (see section 6) been performed in the SQUID case. Rather, what has been done is a spectroscopic experiment which measures the static effect of the tunnelling matrix element Δ. Let us generalize the above discussion to allow the external flux to be different from $\varphi_0/2(\Phi_{ext} = \varphi_0/2 + \delta)$; then the effective two-state Hamiltonian in the $|L\rangle$, $|R\rangle$ representation has (for the simple 'model' case considered) the more general form

$$H_{eff} = \begin{pmatrix} \epsilon/2 & \Delta/2 \\ \Delta/2 & \epsilon/2 \end{pmatrix}, \tag{5.21}$$

where the 'bias' ϵ is to lowest order in δ simply $\varphi_0\delta/L$. The eigenstates of 5.21) are linear superpositions of $|L\rangle$ and $|R\rangle$:

$$\psi_+ = \cos\frac{\theta}{2}|L\rangle + \sin\frac{\theta}{2}|R\rangle \qquad E_+ = -E/2$$

$$\psi = \sin\frac{\theta}{2}|L\rangle - \cos\frac{\theta}{2}|R\rangle \qquad E_+ = +E/2 \tag{5.22}$$

where

$$\tan\theta \equiv \Delta/\epsilon, \qquad E \equiv (\epsilon^2 + \Delta^2)^{1/2} \tag{5.23}$$

and so the splitting is

$$\delta E(\epsilon) = (\epsilon^2 + \Delta^2)^{1/2}. \tag{5.24}$$

Note in particular that in the limit of zero bias the levels are symmetric and antisymmetric combinations of $|L\rangle$ and $|R\rangle$ and are split by an energy Δ. Thus, the energy levels as a function of bias E have the typical 'avoided-crossing' form shown by the solid curve in figure 3. If on the other hand we were to apply the hypothesis of macrorealism, namely that at all times the system is either definitely in $|L\rangle$ or definitely in $|R\rangle$, then the observed energies would be simply the diagonal elements of H_{eff}, i.e. $\pm\epsilon$ as shown by the dashed curves in figure 3 and one would have for the splitting

$$\delta E(\epsilon) = |\epsilon|. \tag{5.25}$$

Thus, a measurement of the splitting as a function of bias can distinguish the two hypotheses.

In practice, the measurement of the splitting is made spectroscopically[32], by adding to the dc external flux Φ_{ext} a small ac component $\delta\Phi_{ext}(t) = \lambda\cos\omega t$: thus, the bias ϵ is time

[32] Note that if the initial state were indeed a mixture of $|L\rangle$ and $|R\rangle$ one would not expect any absorption to occur.

dependent and equal to $\epsilon_0 + \epsilon_1 \cos \omega t$. One then seeks a resonance in the absorption of the ac field as a function of ω, and if such is seen, identifies it with the level splitting. Note that, in strong contrast to the case of ferritin (subsection 5.2), one is here dealing with a 'time ensemble' i.e. a single physical system subjected to repeated probing. This means that only a single microwave photon is absorbed at each 'event', and consequently it is not in practice feasible to measure the absorption by the standard techniques (change of Q-factor of the resonant circuit providing the microwaves etc); rather, one must detect its effect by a more indirect method (see below). It should be noted that the 'intrinsic' efficiency of the spectroscopic method is highest close to the avoided crossing (zero bias) and falls off sharply for $\epsilon \gg \Delta$, since then the two states are nearly localized in the individual wells and a perturbation which is diagonal in this basis has difficulty in coupling them.

Conceptually, both of the recent experiments are close to the above model, but there are some differences in detail. In the Delft experiment [48] the SQUID ring contains not one but three junctions, and moreover its self-inductance is very small, so $\beta_L \ll 1$. The detailed form of the potential energy $U(\Phi)$ as a function of flux is thus different from (5.13), but it preserves the crucial feature, namely that at external flux $\frac{1}{2}\varphi_0$ there are two macroscopically distinct 'ground states', corresponding to clockwise and anticlockwise current flow, which are separated by a barrier which is high enough to be classically impenetrable[33] but not so high that quantum tunnelling through it is negligible. The way in which the resonant absorption of microwave radiation is detected in this experiment is interesting: the flux through the loop is continuously monitored (actually by a second SQUID, of the dc type). Suppose one has a finite, say negative, value of ϵ; then the system tends to be localized more in the $|L\rangle$ well[34], and the average value of the flux is somewhat more than $(1/2)\varphi_0$. This situation persists in the presence of a microwave field, provided that the latter is off-resonance with the level splitting E. However, when it is on resonance it can induce transitions into the higher state, which is localized more in the $|R\rangle$ well, so the measured flux decreases. It is evident that this method of detecting the resonance frequency fails when one is exactly at zero bias and the two states have equal average flux, but one can get meaningful data quite close to this point; see figure 3(a) of [48].

At first sight the above method of detection of QIMDS seems to violate a fundamental principle of quantum measurement theory: should not continuous measurement of the flux value automatically destroy the possibility of superposition of different values? As emphasized by the authors, the reason that it does not is that the measurement is very 'weak' and the data are obtained only by statistical averaging over a large sample. (We recall that a similar consideration applied to the Cooper-pair tunnelling measurements of [28].)

The Stony Brook experiment [49] was on a SQUID of the 'model' type discussed above, so the form of $U(\Phi)$ is given by equation (5.13). However, rather than looking for the 'avoided crossing' of the ground state close to zero bias, the authors apply a fairly large bias and look for an avoided crossing of a pair of excited states, which except close to the crossing point are predominantly localized in one well or the other. Again, microwave radiation is applied to detect the level splitting, but in this case the frequency is held constant and the height and shape of the potential barrier are varied (this is done by variation of the critical current of the junction, which is actually two junctions in parallel, by an auxiliary field applied through the loop between them). As in the Delft experiment, the technique for detecting the absorption is indirect: one starts with the system known to be localized in the upper (say L) well (the

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[33] This conclusion needs a careful accounting for the zero-point energy associated with degrees of freedom 'transverse' to the principal (tunnelling) one; see footnote 12 of [48].

[34] Although it is not clear that the distribution between the two states is the thermal equilibrium one [48], this conclusion follows directly from the measured flux.

barrier is sufficiently high that the probability of tunnelling out of this (metastable 'ground state' is negligible). When radiation is applied, at the resonant frequency, the system can make a transition into an excited state which has an appreciable amplitude in the L-well. If one is near the point at which this state is (in the absence of tunnelling) degenerate with an excited state in the R-well, the system can make a tunnelling transition into this R-well excited state and then decay to the R-well ground state. That it has done so is detected by measuring, just as in the Delft experiment, the average flux through the system loop. Note that, in contrast to the Delft case, the sensitivity in the SUNY experiment is maximum at the crossing point.

It should be emphasized that although in both the SUNY and the Delft experiments the fact that the system makes tunnelling transitions through the classically impenetrable barrier is an essential ingredient in the detection technique, in neither one is the existence of such transitions the principal evidence adduced for QIMDS; rather, the latter is in both cases inferred from the dependence of the energy splitting E on bias ϵ. In both cases the data fit the standard quantum formula (5.24) satisfactorily and are clearly inconsistent with the macrorealistic form (5.25). Needless to say, the absorption peaks in each case are not delta-functions but have appreciable width, which presumably comes from the various sources of dissipation in the SQUID and the associated circuitry; the details of this are an interesting question [50], but one which there is no space to discuss here.

Now let us turn to the question of how 'macroscopic' the superpositions for which those two experiments give evidence are. First, a note on the physical size of the SQUID loops involved: this is approximately 5 μm $\times$ 5 μm for the Delft experiment and approximately 140 μm $\times$ 140 μm for the SUNY one. However, this size is not itself particularly important (in this sense a neutron interferometer is much more 'macroscopic'!) As regards the extensive difference Λ, the most obvious extensive variable to consider is the total magnetic moment; the difference in this quantity between the L- and R-states is about 10^{10} μ_B in the SUNY experiment and about 10^6 in the Delft one. What about the disconnectivity D? The difference in total flux Φ between the two states is close to φ_0 for the Delft experiment, and greater than $\frac{1}{4}\varphi_0$ in the SUNY experiment. A conservative definition of D would then be that it is the order of the number of pairs whose physical state is essentially orthogonal in the two branches[35]— that is, the number which are within a London penetration depth of the surface and actually carry the physical current; it is clear that for the Delft experiment this is precisely of the order of the angular momentum divided by $\hbar$, which within factors of order unity is evidently the same as Λ. For the SUNY experiment, where the single-pair states in the two branches are not close to orthogonal, the value of D could depend on the details of the definition, but most reasonable definitions would make it of the same order. Thus, for the SUNY SQUID experiment both measures of QIMDS, Λ and D, are of order 10^{10}—in each case several orders of magnitude larger than for any other system in which the phenomenon has been reported.

Finally, I should like to emphasize that while there are certainly features (such as the details of the origin of the observed linewidths) of both the SUNY and the Delft experiments which are not completely understood, there is as far as is known at the moment no feature of either experiment which suggests that the interpretation in terms of QIMDS is incorrect.

6. Where do we go from here?

Let us take stock. The original motivation for the programme to search for QIMDS (at least in the minds of many people, including the present author) was to obtain experimental input to the

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[35] It is a moot point whether one should count the formal orthogonality, according to equation (5.11), of the electrons in the bulk of the ring (i.e. beyond the penetration depth); if one did so, the resulting value of D would be somewhat (but not much) larger.

quantum measurement paradox by extending tests of the predictions of QM from the atomic level to that of the everyday world. Note that I say 'everyday world' and not 'macroscopic level': from the point of view of the measurement paradox, it would be of little use to know that QM applies at the level of galaxies, if we could not similarly be sure that it applies to the objects of our immediate experience! Now, while the concept of an 'everyday' object is of course ill-defined and subjective, it would perhaps not be unreasonable to specify that the number of 'elementary' particles comparing a typical such object (and hence the typical values of the measures Λ and D associated with quantum superpositions of the 'significantly' different states of it) is of order 10^{24}. We have seen that recent experiments push tests of QM to the level of 10^3–10^6 in the case of molecular diffraction, biomagnetic and quantum-optical systems, and to the level of 10^{10} in the case of Josephson devices; thus, we can say that on a logarithmic scale we have come about 40% of the way from the atomic level to that of the everyday world. (If that does not sound particularly impressive, recall that until quite recently we had no evidence for QIMDS in any system with D in double figures, i.e. we were <4% of the way!)

Perhaps the most obvious extension of existing work would be that suggested in [29], namely to push experiments on the diffraction of complex molecules in the direction of even heavier objects, such as large biological molecules and eventually small viruses. In a similar manner, there seems no obvious *a priori* objection to extending the SQUID experiments from rings of the size of these used in the existing experiments (a few microns) to much larger sizes, perhaps of the order of 1 cm[36], which would thus correspond to values of Λ and D of the order of 10^{16}. Certainly, the further one goes in this direction the more severe the effects of decoherence are (just as the quantum measurement literature has always claimed) and thus the more difficult one may expect it to be to display the effects of QIMDS, and in addition, in the molecular-diffraction case, the finer the spatial resolution needed; but there seems no obvious *a priori* reason why these difficulties cannot be resolved with the help of improved technology. (For example, in the case of molecular diffraction, the effects of the decoherence due to interaction of the internal degrees of freedom with the black-body radiation field could be drastically reduced if a practical means of preparing the beam at temperatures appreciably lower than the current 900–1000 K could be found.)

However, one might in any case question whether this is the most profitable direction to take at this point. The measurement paradox after all has to do in the last resort with the appearance of violent conflict between the ideas of QM and what we think we know from our immediate conscious experience, so perhaps it would be more useful to move in the direction of the latter. In this spirit it has been suggested [53], for example, that one might try to look for evidence of superposition of the different (though perhaps not 'macroscopically' different) states of the rhodopsin molecule in the human eye which are believed to correspond to the detection or non-detection of a small number of photons (see also [54]). In a context closer to that of the existing experiments, let us suppose that the molecular-diffraction experiments progress to the point where interference of two different centre-of-mass states of, say, a haemoglobin molecule can be detected. At this point, rather than going on to try to observe similar centre-of-mass interference of (say) small viruses, it seems to me that it would be more instructive to try to think of ways of displaying interference of two different internal states of the haemoglobin with different biological functionalities. Certainly, either of these

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[36] For completeness, it should be mentioned that there already exists a series of papers in the literature which claims to see effects of QIMDS in SQUID rings of this size: see [51], and earlier work cited therein. However, it is probably fair to say that the interpretation of the raw data espoused in these papers has not found widespread acceptance in the relevant community. For a critique of some of this work; see [52]; note that the latter does not endorse the claim of QIMDS on the scale of the ∼1 cm ring, only (possibly) a much weaker claim.

proposals raises very severe practical difficulties, since to have maximum significance in the context of the measurement problem the experiments would have to be conducted under the conditions characteristic of biological functionality, in particular with the effective temperature characterizing the internal molecular states at or near room temperature, and the effects of decoherence under such conditions may be expected to be extremely severe. Nevertheless, the whole history of this subject has shown that naive estimates of the effects of decoherence have almost invariably erred on the side of pessimism, sometimes spectacularly so, and it may not be totally ridiculous to hope that this trend will continue in the future.

There is, however, a quite different direction in which it is desirable, and quite possibly feasible, to extend existing experiments. In the language of section 4, we have now completed (at least at the level of SQUID rings) stage 2 of the programme; that is, we have tested the predictions of standard QM, in particular the predictions for the occurrence and effects of QIMDS, and found them to be in agreement with experiment. From the point of view of the strict logic of science, however, this observation does not prove that QM is the correct theory at this level. Indeed, if one accepts the 'falsificationist' position as expounded for example in [55], no finite set of experiments could ever do this; the best that we can ever do is to establish the falsity of a class of theories which are alternative to QM. If that class is sufficiently large, and *a priori* plausible, then establishment of its falsity is itself a worthwhile goal. It is, of course, just such a line of reasoning which motivated the series of 'Bell's-theorem' tests of objective local theories (see section 1) over the last 30 years; when these were initiated, it was only known that QM gave predictions consistent with experiment for distant measurements on correlated photon pairs, while as a result of the experimental programme we can now make the much stronger statement that (with of course the necessary caveats) the whole class of objective local theories is experimentally excluded. In the context of present interest, we are currently at the stage corresponding, in the Bell's-theorem case, to the situation in 1970 (i.e. 'stage 2'); can we go on to stage 3—that is, exclude the whole class of macrorealistic theories?

Before addressing this question as such, let me briefly discuss the situation with regard to the particular member of that class which is best developed in its detailed predictions, namely the GRWP theory (see section 3). With the fundamental parameters a and λ given their currently favoured values, namely 10^{-5} cm and 10^{-16} s^{-1}, this theory is probably not excluded by the existing molecular-diffraction experiments, since while the spatial separation of the two branches of the beam is (just) large enough to trigger the reduction process, the relatively short transit time and fairly small value ($\sim 10^3$) of N mean that the reduction would be negligible at the time when the molecules hit the final screen (cf [13], section 1). The GRWP theory is also not refuted by the SQUID experiments in their current form, principally because the trigger for reduction in this theory is a separation of the centre-of-mass in the two superposed branches, and no such separation occurs in the SQUID experiment as currently conducted. In principle, it would be possible to extend either class of experiment so as to exclude the GRWP theory (or, of course, alternatively to exclude QM!): in the molecular-diffraction case by increasing N and slowing the beam, and in the SQUID case by, for example, an appropriate coupling[37] of the flux through the SQUID to a small magnet suspended on an elastic thread (so that the states L and R correspond to different spatial positions of the magnet's centre of mass).

If one wishes to test the predictions of the whole class of macrorealistic theories against those of QM, an appropriate experiment has been suggested in [15] and discussed in more detail e.g. in [22]. One first needs to define the notion of a 'macrorealistic' theory more precisely: as in the cited reference, I shall define it by the conjunction of three postulates, namely:

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[37] Note, however, that this would, by the usual Franck–Condon effect, tend to drastically reduce the tunnelling splitting Δ, and this effect would have to be compensated.

(1) *Macrorealism per se.* A macroscopic object which has available to it two or more macroscopically distinct states is at any given time[38] in a definite one of those states.
(2) *Non-invasive measurability.* It is possible in principle to determine which of these states the system is in without any effect on the state itself or on the subsequent system dynamics.
(3) *Induction.* The properties of ensembles are determined exclusively by initial conditions (and in particular not by final conditions).

While it has occasionally been argued (e.g. [56]) that the postulate (2) is not a necessary ingredient of a theory which can reasonably be called 'macrorealistic', the present author's view is that it is so natural a corollary of (1) that the latter is virtually meaningless in its absence: see [15, 22]. On the question of postulate (3) (which has not been seriously challenged in the literature, at least to my knowledge), see [57]).

Consider now a physical system possessing some observable Q which, whenever measured, is found to take a value ± 1 (only), and consider a series of runs starting from identical initial conditions such that on the first set of runs Q is measured (only) at times t_1 and t_2, on the second only at times t_2 and t_3, on the third at t_3 and t_4 and, finally, in the fourth set, only at t_1 and t_4 ($t_1 < t_2 < t_3 < t_4$). Directly from the results of the measurements, one establishes the correlations $C_{ij} \equiv \langle Q(t_i) Q(t_j) \rangle$. Then, by a straightforward adaptation of standard arguments for one of the 'CHSH inequalities' [58] in the Bell's-theorem area, one establishes [15] that any macrorealistic theory predicts the inequality

$$K \equiv C_{12} + C_{23} + C_{34} - C_{14} \leqslant 2 \qquad \text{(MR)} \qquad (6.1)$$

for any values of the t_i. Note that the only requirement for the proof, other than the postulates defining macrorealism themselves, is that Q takes only the values[39] ± 1.

The punch line, as one might guess from the analogous result in the Bell's-theorem context, is that under certain conditions the predictions of QM violate[40] inequality (6.1).

Consider for example a symmetric two-well system of the type formed by a SQUID with external bias $\varphi_0/2$ (section 5) and define $Q(t)$ to be the 'coarse-grained' flux; i.e., the measured value of Q is defined to be $+1$ if the flux is measured and found to be (sufficiently close to) $\varphi_0/2$, and -1 if it is (sufficiently close to) $-\varphi_0/2$. Take the times t_i to satisfy the condition

$$t_2 - t_1 = t_3 - t_2 = t_4 - t_3 = \pi/(4\Delta)$$

where Δ is the tunnelling splitting of the two-state system so formed. Then in the total absence of dissipation (decoherence), the prediction of QM is

$$K = 2\sqrt{2} \qquad \text{(QM, ideal)} \qquad (6.2)$$

clearly violating the MR inequality (6.1). This result, of course, is not in itself of much interest, since the total neglect of dissipation is unrealistic. However, in the specific case of a SQUID system, detailed calculations of the effect of decoherence have been carried out using the 'classical–quantum correspondence method' described in section 4 (see e.g. [60]) and the result is that under not obviously unattainable conditions one predicts

$$K > 2 \qquad \text{(QM, realistic)} \qquad (6.3)$$

still in conflict with the MR result (6.1). Thus, an appropriate experiment of this type can distinguish unambiguously between the predictions (at the level of SQUIDs) of QM and those of an arbitrary macrorealistic theory.

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[38] Except for possible short 'transit times' which can be allowed for in the derivation of the inequality below (see [15]).
[39] But a small probability of obtaining values other than these can be accommodated in the proof; see [15].
[40] That Bell-type inequalities may be profitably used on a single system had been earlier noted by Home and Sengupta [59].

The conditions on the degree of decoherence etc are considerably more stringent for such a 'real-time' experiment than for the spectroscopic experiments so far conducted; in particular, the effects of the external circuits necessary to perform the measurements of Q need to be considered in detail, see [61, 62]. Recently, a detailed analysis of the feasibility of this experiment using currently available technology has been carried out in chapter 4 of [50], and the conclusion (at least in the present author's reading!) is that while it is very difficult, it is not so obviously impossible that one should not try to work towards it (cf also [61, 62]).

A major external impetus towards pushing this program forward has developed in the last few years from a direction which at the time of its initiation could not have been anticipated, namely the hope of using circuits such as SQUID rings as elements ('qubits') in a solid-state quantum computer [63]. Indeed, as this article goes to press, there has been a very exciting development in this area: the group at Saclay has reported [64] experiments on a Josephson circuit which observe coherent oscillations with a quality factor of 25 000. The circuit in question is a 'hybrid' one, which combines the principles of the Cooper-pair box and the single-junction SQUID, and the analysis is therefore somewhat more complicated than for the simple SQUID; in fact, the above very high quality factor, was observed only under conditions such that the two states involved do not differ appreciably in any macroscopic property. However, in the present context the salient result is that a 'large' ($\sim$50) value of the factor is still observed even when the two states are indeed 'macroscopically distinct' (while both the linear dimensions and the circulating currents are smaller than in the SUNY experiments, the values of lambda and D are still $\sim 10^4$–10^5, and there seems no obvious reason that they could not be increased). In view of the crucial significance of the two-time correlation experiment discussed above for our whole view of everyday (or at least SQUID-level!) 'reality', it is to be hoped that it will be attempted on such a system in the near future.

Acknowledgments

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