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Entropy and Irreversibility for a Single Isolated Two Level System: New Individual Quantum States and New Nonlinear Equation of Motion

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We propose a new nonlinear equation of motion for a single isolated two-level quantum system. The resulting generalized two-level quantum dynamical theory entails a new alternative resolution of the long-standing dilemma on the nature of entropy and irreversibility. Even for a single isolated degree of freedom, in addition to the individual mechanical states for which all the results of conventional quantum mechanics remain valid, our theory implies the existence of new nonmechanical individual quantum states. These states have nonzero individual entropy and, by virtue of a constant-energy, internal redistribution mechanism, relax irreversibly toward stable equilibrium. We discuss the possibility of an experimental verification of these conclusions by means of a high-resolution, essentially single-particle, magnetic-resonance experiment.

1. INTRODUCTION

The long-standing dilemma on the nature of entropy and irreversibility still lacks a universally accepted resolution,[2] in spite of a century of scientific efforts. As stated in a recent review by Wehrl (1978): "There are many opinions and proposals for a solution to this problem; however, none of them seems to be completely satisfactory." The purpose of this paper is to present a novel nonlinear equation of motion for a single two-level quantum system that was proposed by the author[3] in an effort to attempt a satisfactory resolution of the irreversibility dilemma.

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[2]For a recent critical review of the different schools of thought, see Park and Simmons (1983).

[3]See Beretta (1981). The general form of the new equation of motion for a single constituent of matter is presented in Beretta *et al.* (1984) and in Beretta (1985).

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Our approach differs essentially from the traditional where entropy, irreversibility, and the laws of thermodynamics are invariably regarded as statistical, macroscopic, or phenomenological concepts with no fundamental counterpart in the microscopic reality. Indeed, our underlying premise is that the gap between mechanics and thermodynamics can be conceivably bridged without resorting to any statistical or information-theoretic reasoning, without hinging on the distinction between microscopic and macroscopic reality, and without regarding the laws of thermodynamics as simply "phenomenological."

For definiteness, we discuss only the simplest case of a single two-level quantum system. Our proposal, based on the two fundamental hypotheses presented in Section 2, provides a quite unconventional, but logically coherent, new resolution of the irreversibility dilemma consistent with the declared premise. Irreversibility emerges as a manifestation of an inherent energy-conserving relaxation mechanism implied by the postulated new equation of motion, even in the absence of any form of interaction of the system with any other system, lattice or "heat bath," i.e., even for a strictly isolated two-level system.

In addition to all the results of conventional quantum mechanics which hold as special cases of our theory, the new equation of motion implies the existence of inherent relaxation effects that should be in principle experimentally verifiable. For this purpose, we study the dynamics of a single spin-1/2 system in an external time-dependent magnetic field and propose that the predicted single-particle relaxation effect may be verified with a high-resolution magnetic-resonance experiment on a beam of spin-1/2 particles of very low intensity (essentially single particle).

The paper is organized as follows. Section 2 presents the two new fundamental postulates of our theory. Section 3 discusses the general properties of the new nonlinear equation of motion that we propose for a single isolated two-level system. Section 4 presents special classes of exact and approximate solutions of the equation of motion. Section 5 proposes one method to verify experimentally the validity of our two postulates and Section 6 gives conclusions.

2. NONIDEMPOTENT QUANTUM STATES AND NONLINEAR EQUATION OF MOTION

Our first fundamental hypothesis is due to Hatsopoulos and Gyftopoulos (1976). They proposed that, in addition to the individual quantum states conceived of within conventional quantum mechanics and represented mathematically in terms of idempotent state operators,[4] a single strictly isolated (i.e., noninteracting and uncorrelated) system admits also of individual quantum states that must be represented by nonidempotent state operators. State operators ρ have the same mathematical properties[4] as the statistical or density operators considered in traditional (von Neumann) quantum statistical mechanics,[5] but acquire in our theory an entirely different physical meaning. A state operator ρ represents an individual quantum state of the single strictly isolated system. It does not represent the index of statistics from a generally heterogeneous ensemble of identical systems.

As shown by Hatsopoulos and Gyftopoulos (1976), the only trace functional of the state operator ρ that can represent the physical observable entropy is $s(\rho) = -k \operatorname{Tr} \rho \ln \rho$. The nonlinear state functional $s(\rho)$ is defined for all state operators, idempotent and nonidempotent. It represents the individual entropy of the single strictly isolated and uncorrelated system. It does not represent a measure of statistical or information-theoretic uncertainty.

In summary, with the Hatsopoulos–Gyftopoulos fundamental hypothesis, we conceive of a larger set of individual quantum states of uncorrelated systems than in conventional quantum mechanics. A single isolated system may be found not only in a quantum mechanical state $[\rho^2 = \rho, s(\rho) = 0]$ but also in a nonmechanical quantum state described by a nonidempotent state operator ρ for which the entropy functional is nonzero.

Our next step is to describe the time evolution of all the individual quantum states of a single isolated two-level system. For quantum mechanical individual states, the dynamical law is the Schrödinger equation of motion or, in terms of state operators, the von Neumann equation of motion. But, for the additional nonmechanical individual quantum states that we have postulated, the dynamical law cannot be "derived" from conventional quantum mechanics, simply because that theory cannot even conceive of such individual states. We must therefore augment the dynami-

[4]A state operator ρ is a linear, self-adjoint, nonnegative-definite, unit-trace operator on the Hilbert space of the system, i.e., $\rho^\dagger = \rho$, $\rho \geq 0$, $\operatorname{Tr} \rho = 1$. Every idempotent state operator $(\rho^2 = \rho)$ is a projector onto the one-dimensional span of a vector ψ called the quantum mechanical state vector of the system. For a single strictly isolated (i.e., noninteracting and uncorrelated) system, conventional quantum mechanics conceives only of individual quantum states that can be represented by a state vector or, equivalently, by an idempotent state operator.

[5]On the conceptual problems arising from the traditional use of the statistical operator, see Park (1968).

cal postulate with a new equation of motion, consistent with the Schrödinger equation for idempotent state operators.

Our second fundamental hypothesis is the following equation of motion (Beretta, 1981; Beretta et al., 1984; Beretta, 1985) that we propose for a single isolated two-level system

$$\frac{d\rho}{dt}+\frac{i}{\hbar}[H,\rho]=\begin{cases} -\dfrac{1}{\tau}\dfrac{\begin{vmatrix} \rho\ln\rho & \rho & \frac{1}{2}\{H,\rho\} \\ \mathrm{Tr}\,\rho\ln\rho & 1 & \mathrm{Tr}\,\rho H \\ \mathrm{Tr}\,\rho \mathrm{H}\ln\rho & \mathrm{Tr}\,\rho \mathrm{H} & \mathrm{Tr}\,\rho H^2 \end{vmatrix}}{\mathrm{Tr}\,\rho H^2-(\mathrm{Tr}\,\rho H)^2}, & \text{if } \rho^2\neq\rho \quad (1a) \\ 0, & \text{if } \rho^2=\rho \quad (1b) \end{cases}$$

where H is the Hamiltonian operator and τ is an inherent internal-redistribution time constant of the system. For the same reason why the dynamical law for nonmechanical states cannot be "derived" from mechanics, the value of the time constant τ cannot be obtained other than by analysis of experimental data. Equation (1) has been "invented," not "derived." Its adoption is justified only insofar as its consequences are consistent with our declared premises, in particular, with the laws of mechanics for idempotent individual states and with the laws of thermodynamics for all individual states.

For example, we will see that a consequence of equation (1) is a statement that the entropy of the strictly isolated two-level system is constant for all the mechanical states $[s(\rho)=0]$ and for all the equilibrium states (there is one equilibrium state for each initial value of the mean energy), but it is strictly increasing in time for all other nonmechanical states. Again, consistently with the fact that it describes the time evolution of an isolated system, equation (1) conserves the mean energy.

3. PROPERTIES OF THE NEW EQUATION

On the two-dimensional Hilbert space of the two-level system, we introduce the 3-vector $\mathbf{R}=(R_1, R_2, R_3)$ of spin operators which obey the commutation rule $[R_j, R_m]=i\varepsilon_{jmn}R_n$ and may be expressed in terms of the lowering and raising operators $b=R_1-iR_2$ and $b^\dagger=R_1+iR_2$. A state operator $\rho(t)$ may then be represented as

$$\rho(t)=\tfrac{1}{2}I+r_1(t)R_1+r_2(t)R_2+r_3(t)R_3=\tfrac{1}{2}I+\mathbf{r}(t)\cdot\mathbf{R} \quad (2a)$$

$$=\tfrac{1}{2}I+\alpha^*(t)b^\dagger+\alpha(t)b+r_3(t)\mathbf{R}_3 \quad (2b)$$

where the 3-vector $\mathbf{r}=(r_1, r_2, r_3)$ of real scalars and the complex scalar α satisfy the condition

$$r=|\mathbf{r}|=(r_1^2+r_2^2+r_3^2)^{1/2}=(4\alpha^*\alpha+r_3^2)^{1/2}\leq 1 \quad (2c)$$

Geometrically, the set of state operators is isomorphic with a closed spherical domain of unit radius (the Bloch sphere) in an auxiliary three-dimensional space with orthogonal coordinates r_1, r_2, and r_3. Each point $\mathbf{r}$ in the Bloch sphere represents, via relation (2a), a state operator. A state operator is idempotent ($\rho^2=\rho$) if and only if $r=1$. Thus, all the state operators conceived of within conventional quantum mechanics (for a single strictly isolated two-level system) lie on the surface of the Bloch sphere. The Hatsopoulos-Gyftopoulos hypothesis extends the domain of conceivable individual states of uncorrelated two-level systems to the whole volume inside the Bloch sphere, including state operators for which $\rho \neq \rho^2$ and $r<1$.

A general Hamiltonian operator corresponding to the energy relative to a point midway between the two energy levels of the isolated two-level system may be represented as

$$H=\hbar\Omega_0(\Lambda_1 R_1+\Lambda_2 R_2+\Lambda_3 R_3)=\hbar\Omega_0\mathbf{\Lambda}\cdot\mathbf{R} \tag{3a}$$

$$=-\tfrac{1}{2}\hbar\Omega(\varepsilon b^\dagger+\varepsilon^* b)+\hbar\omega_0 R_3 \tag{3b}$$

where $\hbar$ is the reduced Planck constant, $\mathbf{\Lambda}=(\Lambda_1,\Lambda_2,\Lambda_3)$ is a unit-norm 3-vector of real scalars ($|\mathbf{\Lambda}|=\mathbf{\Lambda}\cdot\mathbf{\Lambda}=1$), ε is a complex scalar with $\varepsilon^*\varepsilon=1$, $\omega_0=\Omega_0\Lambda_3$ is the transition frequency between the two levels and $\Omega_0^2=\Omega^2+\omega_0^2$.

If $\mathbf{r}=\pm\mathbf{\Lambda}$, then $\rho_\pm=\rho_\pm^2=P_{\psi_\pm}$ where $\psi_\pm$ are the eigenvectors of the Hamiltonian operator H. According to equation (1), and consistently with conventional quantum mechanics, the two pure mechanical states $\rho_\pm$ are equilibrium states. Assuming from now on that $\mathbf{r}\neq\pm\mathbf{\Lambda}$, after lengthy but straightforward manipulations (outlined in Appendix A), using relations (2) and (3) for operators ρ and H in equation (1) with $\mathbf{r}\neq\pm\mathbf{\Lambda}$, we find the following forms of the new equation of motion:

$$\frac{d\mathbf{r}}{dt}-\Omega_0\mathbf{\Lambda}\times\mathbf{r}=-\frac{1}{\tau}K(\mathbf{r})[\mathbf{r}-(\mathbf{\Lambda}\cdot\mathbf{r})\mathbf{\Lambda}] \tag{4a}$$

$$=-\frac{1}{\tau}K(\mathbf{r})[\mathbf{\Lambda}\times\mathbf{r}\times\mathbf{\Lambda}] \tag{4b}$$

or

$$\frac{d\alpha}{dt}+i\omega_0\alpha+\frac{i\Omega\varepsilon r_3}{2}=-\frac{K(\alpha,r_3)}{\tau\Omega_0^2}\left[\omega_0^2\alpha+\frac{\omega_0\Omega\varepsilon r_3}{2}+\frac{\Omega^2\varepsilon(\varepsilon^*\alpha-\varepsilon\alpha^*)}{2}\right] \tag{5a}$$

$$\frac{dr_3}{dt}+i\Omega(\varepsilon^*\alpha-\varepsilon\alpha^*)=-\frac{K(\alpha,r_3)}{\tau\Omega_0^2}[\Omega^2 r_3+\omega_0\Omega(\varepsilon^*\alpha+\varepsilon\alpha^*)] \tag{5b}$$

where

$$K(\mathbf{r}) = \frac{f(r)}{1-(\mathbf{\Lambda}\cdot\mathbf{r})^2} = K(\alpha, r_3) = \frac{f(r)}{1-[\omega_0 r_3 - \Omega(\varepsilon\alpha + \varepsilon^*\alpha^*)]^2/\Omega_0^2} \tag{6}$$

$r = |\mathbf{r}|$, and

$$f(r) = \begin{cases} 1, & \text{if } r = 0 \\ \dfrac{1-r^2}{2r}\ln\dfrac{1+r}{1-r}, & \text{if } 0 < r < 1 \\ 0, & \text{if } r = 1 \end{cases} \tag{7}$$

As shown in Appendix B, for every initial state $\mathbf{r}_0$ in the Bloch sphere ($\mathbf{r}_0 \leq 1$), the new equation of motion admits of one and only one solution $\mathbf{r}(t)$ with $\mathbf{r}(0) = \mathbf{r}_0$ which lies entirely in the Bloch sphere for $-\infty < t < +\infty$. Thus, let $\mathbf{r}(t)$ be a solution with $\mathbf{r}(0)$ in the Bloch sphere, i.e., with $r(0) \leq 1$. From equation (4), we find that

$$\frac{d}{dt}\operatorname{Tr} H\rho = \frac{d}{dt}\frac{1}{2}\hbar\Omega_0\mathbf{\Lambda}\cdot\mathbf{r} = \frac{1}{2}\hbar\Omega_0\mathbf{\Lambda}\cdot\frac{d\mathbf{r}}{dt} = 0 \tag{8}$$

and, therefore, we conclude that the mean energy Tr $H\rho$ is a constant of the motion for the isolated two-level system, consistently with the first law of thermodynamics. Every solution lies entirely on a constant mean energy plane orthogonal to $\mathbf{\Lambda}$ and at distance $|\mathbf{\Lambda}\cdot\mathbf{r}|$ from the origin.

Again from equations (4), (6), and (7), we find that

$$\frac{dr}{dt} = -\frac{1}{\tau}\frac{r^2 - r_e^2}{1-r_e^2}\frac{1-r^2}{2r^2}\ln\frac{1+r}{1-r}\begin{cases} =0, & \text{if } |r_e| < r = 1 \\ <0, & \text{if } |r_e| < r < 1 \\ =0, & \text{if } |r_e| = r < 1 \end{cases} \tag{9}$$

where $r_e = \mathbf{\Lambda}\cdot\mathbf{r}$. As shown in Appendix B, a consequence of relation (9) is that the solution $\mathbf{r}(t)$ remains within the Bloch sphere at all times. In terms of state operators, this implies that the solution $\rho(t)$ remains at all times within the set of state operators and, therefore, equation (1) is a valid evolution equation.

A special class of solutions of equation (4) is such that $r(t) = 1$ at all times. In this case, the right-hand side of equation (4) vanishes for every t [$f(1) = 0$, equation (7)] and thus $\mathbf{r}(t)$ satisfies the differential equation

$$\frac{d\mathbf{r}}{dt} = \Omega_0\mathbf{\Lambda}\times\mathbf{r} \tag{10}$$

For this class of solutions, the vector $\mathbf{r}$ remains on the unit-radius surface of the Bloch sphere and precesses around $\mathbf{\Lambda}$ at the Larmor angular frequency Ω_0. In terms of state operators, $\rho(t)$ remains idempotent at all times and satisfies the operator form of the Schrödinger equation of motion [equation (1b)]. We conclude that all the motions contemplated by conventional quantum dynamics are special solutions of equation (1).

Next we consider the entropy functional

$$s(\rho) = -k \operatorname{Tr} \rho \ln \rho = -\tfrac{1}{2}k[(1+r)\ln(1+r) + (1-r)\ln(1-r) - \ln 4] \tag{11}$$

which is a monotonic decreasing function of r ranging from $k \ln 2$ to 0 as r ranges from 0 to 1. In the Bloch sphere, concentric spherical surfaces centered at the origin are constant entropy. A direct consequence of relation (9) is that $ds(\rho)/dt \geq 0$. More explicitly,

$$\frac{ds}{dt} = \frac{k}{\tau}\frac{r^2 - r_e^2}{1 - r_e^2}\frac{1 - r^2}{4r}\left(\ln\frac{1+r}{1-r}\right)^2 \geq 0 \tag{12}$$

with strict inequality for $|r_e| < r < 1$. We conclude that the entropy of an isolated two-level system is a nondecreasing function of time, consistently with the second law of thermodynamics. Moreover, for solutions with $r(0) < 1$ the entropy is strictly increasing, i.e., equation (1) implies the existence of irreversible, but energy-conserving relaxation even for a single isolated system. This nonconventional consequence of the proposed equation of motion should be in principle experimentally verifiable (cf. Section 5).

The equilibrium states, for which $d\mathbf{r}/dt = 0$, are only those with $r = r_e$, i.e., with $\mathbf{r}$ parallel to $\mathbf{\Lambda}$. It readily follows from relation (9) that these equilibrium states are stable. Only two equilibrium states are "mechanical" (i.e., idempotent), namely, those with $\mathbf{r} = \pm\mathbf{\Lambda}$. All the others, with $\mathbf{r} = r_e\mathbf{\Lambda}$ and $-1 < r_e < 1$, are nonidempotent individual equilibrium states not contemplated by conventional mechanics that, in terms of state operators, have the form

$$\rho_e = \frac{\exp(-\beta H)}{\operatorname{Tr}\exp(-\beta H)} \tag{13}$$

with

$$\beta = \frac{1}{\hbar\Omega_0}\ln\frac{1 - r_e}{1 + r_e} \tag{14}$$

and, according to the Hatsopoulos–Gyftopoulos hypothesis, represent the thermodynamic equilibrium states of a single isolated two-level system with individual thermodynamic temperature $T = 1/k\beta$ (positive if $r_e < 0$).

To summarize, in the Bloch sphere the solutions of equation (1) describe constant mean energy trajectories of the state point $\mathbf{r}$ with velocity given by two orthogonal components. The first component causes a precession of $\mathbf{r}$ around the vector $\mathbf{\Lambda}$ and is due to the Hamiltonian, or Schrödinger, term in the equation of motion. The second component, whose intensity is a nonlinear function of $\mathbf{r}$ vanishing for $r=1$, causes an attraction, or relaxation, of $\mathbf{r}$ towards the stable equilibrium vector $\mathbf{r}_e = r_e \mathbf{\Lambda}$ and is due to the non-Hamiltonian nonlinear term in the equation of motion. The resulting motion is a simple precession at the Larmor frequency Ω_0 if $r(0)=1$, whereas it is a spiraling relaxation towards the equilibrium vector if $r(0)<1$.

By contrast with the phenomenological descriptions based on the Bloch relaxation equation (Pake, 1973; Schumacher, 1970; Poole and Farach, 1972; Rabi, Ramsey, and Schwinger, 1954; Weger, 1960), the relaxation mechanism that we postulate by adopting equation (1) is not due to any form of coupling between the two-level system and other external degrees of freedom. It is a nonlinear, mean-energy conserving, redistribution mechanism strictly internal and individual to the single strictly isolated system.

4. SOLUTIONS OF THE EQUATION OF MOTION

Equation (4) reduces to the scalar equation

$$\frac{d|\mathbf{r}-\mathbf{r}_e|}{dt} = -\frac{1}{\tau} f(r) \frac{|\mathbf{r}-\mathbf{r}_e|}{1-r_e^2} \tag{15}$$

The nonlinearity of this equation does not allow a general explicit solution. However, for $r_e=0$, it becomes

$$\frac{dr}{dt} = -\frac{1}{\tau} f(r) r \tag{16}$$

or, equivalently,

$$\frac{d}{dt} \ln \frac{1+r}{1-r} = -\frac{1}{\tau} \ln \frac{1+r}{1-r} \tag{17}$$

which integrated from 0 to t yields,

$$\frac{1+r(t)}{1-r(t)} = \left[\frac{1+r(0)}{1-r(0)}\right]^{\exp(-t/\tau)} \tag{18}$$

or, equivalently,

$$r(t) = \tanh\left[\frac{1}{2} \exp\left(-\frac{t}{\tau}\right) \ln \frac{1+r(0)}{1-r(0)}\right] \tag{19}$$

Thus, we found a class of special solutions of the new equation of motion which satisfy initial conditions with $r(0) < 1$ and $r_e = 0$, i.e., initial nonidempotent state operators with zero mean energy.

Next, we consider the limit as $\mathbf{r}(0)$ tends to $\mathbf{r}_e$. Then, we can use the approximation $f(r) \approx f(r_e)$ and linearize equation (15) to yield

$$|\mathbf{r}(t) - \mathbf{r}_e| \approx |\mathbf{r}(0) - \mathbf{r}_e| \exp(-t/T_R) \tag{20}$$

where, using equation (14), the approximate relaxation time T_R is given by

$$T_R = \tau \frac{1 - r_e^2}{f(r_e)} = -\tau \frac{r_e}{\frac{1}{2}\hbar\Omega_0\beta} = \tau \frac{\tanh(\frac{1}{2}\hbar\Omega_0\beta)}{\frac{1}{2}\hbar\Omega_0\beta} \tag{21}$$

and, for $\frac{1}{2}\hbar\Omega_0\beta$ sufficiently small (sufficiently high temperature of the stable equilibrium state $\mathbf{r}_e$),

$$T_R = \tau[1 - \tfrac{1}{3}(\tfrac{1}{2}\hbar\Omega_0\beta)^2 + \tfrac{2}{15}(\tfrac{1}{2}\hbar\Omega_0\beta)^4 - \cdots] \tag{22}$$

It is interesting to note that our intrinsic relaxation or redistribution mechanism shows in this approximation an explicit temperature dependence which in principle should be experimentally verifiable, for example as discussed in the next section.

5. A SINGLE-PARTICLE MAGNETIC RESONANCE EXPERIMENT

As a first attempt to search for an experimental verification of our hypotheses and, in particular, of the implications of equation (1), we consider a very-low-intensity (essentially single-particle) beam of spin-1/2 particles entering a suitable magnetic resonance apparatus. During its residence time in the apparatus, each single particle experiences a time-dependent spatially uniform applied magnetic field.

In the laboratory reference frame, we assume that inside the magnetic resonance apparatus the external magnetic field has components given by

$$\mathbf{H}_M(t) = (H_{M1} \cos \omega t, H_{M1} \sin \omega t, H_{M3}) \tag{23}$$

while outside the apparatus, $H_{M1} = 0$. With respect to a reference frame rotating about the third laboratory axis $\mathbf{k}_3$ at the angular frequency ω, the magnetic field has constant components

$$\mathbf{H}_M(t) = (H_{M1}, 0, H_{M3})' \tag{24}$$

The magnetic moment operator vector of the spin-1/2 particle with gyromagnetic ratio γ is given by

$$\mathbf{M} = \gamma\hbar\mathbf{R} \tag{25}$$

and the Hamiltonian operator by

$$H = \mathbf{H}_M(t) \cdot \mathbf{M} = \hbar\Omega(R_1 \cos \omega t + R_2 \sin \omega t) + \hbar\omega_0 R_3 \tag{26}$$

By comparison with equation (3), we find

$$\Omega_0 \mathbf{\Lambda} = (\Omega \cos \omega t, \Omega \sin \omega, \omega_0) = (\Omega, 0, \omega_0)' \tag{27}$$

where $\Omega = \gamma H_{M1}$, $\omega_0 = \gamma H_{M3}$ and $\Omega_0^2 = \Omega^2 + \omega_0^2$.

If d'/dt denotes differentiation with respect to the rotating frame, then $d\mathbf{r}/dt = d'\mathbf{r}/dt + \omega\mathbf{k}_3 \times \mathbf{r}$ and equation (4) becomes

$$\frac{d'\mathbf{r}}{dt} = (\Omega_0 \mathbf{\Lambda} - \omega \mathbf{k}_3) \times \mathbf{r} - \frac{1}{\tau} K(\mathbf{r})[\mathbf{r} - r_e \mathbf{\Lambda}] \tag{28}$$

Owing to the time dependence of the Hamiltonian, the mean energy, $\text{Tr}\, H\rho = \hbar\Omega_0 r_e/2$, changes at a rate given by

$$\frac{dr_e}{dt} = \frac{d}{dt} \mathbf{\Lambda} \cdot \mathbf{r} = \frac{d\mathbf{\Lambda}}{dt} \cdot r = \frac{d'}{dt} \mathbf{\Lambda} \cdot \mathbf{r} = \mathbf{\Lambda} \cdot \frac{d'\mathbf{r}}{dt} = \frac{\omega\Omega}{\Omega_0} r_2' \tag{29}$$

where r_1', r_2', r_3' denote the components of $\mathbf{r}$ with respect to the rotating frame, i.e., $\mathbf{r} = (r_1', r_2', r_3')'$.

To find an approximate solution of equation (28) we will make the following assumptions, which are similar to the so-called "slow-passage" or "steady-state" conditions (see e.g., Pake, 1973) of standard magnetic-resonance configurations:

(1) The relative change in mean energy during the residence time t_{res} in the apparatus is small, i.e., $|t_{\text{res}} \dot{r}_e / r_e| \ll 1$, so that we can consider vector $\mathbf{r}_e$ as slowly varying in the rotating frame.

(2) The spin-1/2 system enters the apparatus in a nonidempotent stable equilibrium state with respect to the outside external magnetic field and $\Omega \ll \omega_0$, so that the vector $\mathbf{r}$ remains close to $\mathbf{r}_e$ and we can consider $K(\mathbf{r}) \approx 1/T_R \approx \text{const}$, as done in equation (20).

(3) The inherent relaxation mechanism maintains an approximately constant magnetization in the rotating frame, so that we can consider $d'\mathbf{r}/dt \approx 0$.

Under these assumptions, the solution of homogeneous equation (28) can be approximated by a slowly varying quasisteady solution. Setting $d'\mathbf{r}/dt = 0$ and considering r_e as constant, equation (28) yields

$$-(\omega_0 - \omega) r_2' - r_1'/T_R = -\Omega r_e / T_R \Omega_0 \tag{30a}$$

$$(\omega_0 - \omega) r_1' - \Omega r_3' - r_2'/T_R = 0 \tag{30b}$$

$$\Omega r_2' - r_3'/T_R = -\omega_0 r_e / T_R \Omega_0 \tag{30c}$$

$$\begin{bmatrix} r_1' \\ r_2' \\ r_3' \end{bmatrix} = \frac{r_e}{\Omega_0[1+\Omega^2 T_R^2 + (\omega_0-\omega)^2 T_R^2]} \begin{bmatrix} \Omega[1+\Omega^2 T_R^2 + (\omega_0-\omega)\omega_0 T_R^2] \\ -\Omega\omega T_R \\ \omega_0[1+(\omega_0-\omega)^2 T_R^2] + (\omega_0-\omega)\Omega^2 T_R^2 \end{bmatrix} \tag{31}$$

Defining the effective relaxation time

$$T_E = \frac{T_R}{(1+\Omega^2 T_R^2)^{1/2}} \tag{32}$$

and the complex susceptibility

$$\chi = \chi' + i\chi'' = \frac{\mathrm{Tr}\,\rho(M_1 - iM_2)}{H_{M1}} = \frac{\gamma^2 \hbar}{2\Omega}(r_1' - ir_2') \tag{33}$$

we obtain

$$\begin{bmatrix} \chi' \\ \chi'' \end{bmatrix} = \frac{\gamma^2 \hbar r_e}{2\Omega_0[1+(\omega_0-\omega)^2 T_E^2]} \begin{bmatrix} 1+(\omega_0-\omega)\omega_0 T_E^2 \\ -\omega T_E^2/T_R \end{bmatrix} \tag{34}$$

and, from equations (29) and (31),

$$-\frac{1}{r_e}\frac{dr_e}{dt} = \frac{\omega^2 \Omega^2 T_E^2 / T_R}{\Omega_0[1+(\omega_0-\omega)^2 T_E^2]} \tag{35}$$

At the frequency $\omega = \omega_0 + 1/\omega_0 T_E^2$, the right-hand side of equation (35) achieves a maximum value $\Omega^2(1+\omega_0^2 T_E^2)/\Omega_0^2 T_R$ which must be much smaller than $1/t_{\mathrm{res}}$ for consistency with our assumption (1).

Compared to the typical dispersion and absorption curves observed in conventional magnetic-resonance experiments under slow-passage or steady-state conditions (see e.g., Pake, 1973), the real and imaginary parts of the complex susceptibility as given by equation (34) present asymmetries with respect to the resonance condition $\omega = \omega_0$. Their shape is determined by the parameter T_E which is in turn related to the unknown internal time constant τ and the individual inverse temperature β via relation (21).

As already discussed, the single-particle relaxation effect predicted here is entirely different from the spin–lattice and spin–spin relaxation effects that dominate in standard magnetic resonance experiments where each single spin-1/2 system is coupled to external degrees of freedom. However, the effect predicted here should be experimentally verifiable with high resolution measurements made on a very diluted beam of spin-1/2 particles so that the only external interactions felt by each single particle are those with the applied magnetic fields.

An experiment on these lines would verify the validity of our two hypothesis, namely, the existence of nonmechanical individual states described by nonidempotent state operators and the new equation of motion describing their irreversible time evolution. The experiment would determine the value of the unknown time constant τ for the spin-1/2 particle or, at least, a lower bound to that value.

6. CONCLUSIONS

We have proposed a new approach towards a satisfactory resolution of the so-called irreversibility dilemma or paradox. Instead of statistical, information-theoretic, macroscopic, phenomenological, or anthropomorphic concepts, we submit that entropy and irreversibility are microscopic physical concepts, in the same sense as energy is a microscopic physical concept, and are defined for each individual physical system, even if composed of a single strictly isolated degree of freedom. To show that a logically consistent approach based on this unconventional premise is feasible, we proposed a generalized quantum dynamical theory for the simplest quantum system, namely, a single isolated two-level system.

In conventional quantum mechanics, the entropy of a single individual strictly isolated system is an undefined concept (entropylike concepts are defined only for the statistical description of ensembles of such systems). In our generalized quantum dynamics, instead, entropy emerges as a physical observable of every single isolated two-level system, and is defined for each of its individual states. We achieve this by adopting the Hatsopoulos-Gyftopoulos postulate that, in addition to all the traditional individual quantum mechanical states (with individual entropy now defined and equal to zero), there exists a broad class of nonmechanical individual quantum states (inconceivable within conventional mechanics and with nonzero entropy).

In conventional quantum mechanics, individual states of a single isolated system evolve in time only along reversible paths (Schrödinger equation of motion). Irreversibility has no place within mechanics. To explain a physical reality dominated by irreversible processes, mechanics is invariably complemented by additional postulates (such as those of the statistical, information-theoretic, macroscopic, or phenomenological approaches reviewed by Park and Simmons, 1983). In our generalized (nonstatistical) quantum dynamics, the additional postulates are the Hatsopoulos-Gyftopoulos hypothesis and the new nonlinear equation of motion proposed by the author. Irreversibility emerges as a microscopic physical phenomenon occurring within the single isolated system in most of its nonmechanical individual states.

The proposed two-level quantum dynamics broadens the quantum mechanical treatment without contradicting any of its successful results. But the generalized theory implies new additional results that are inconceivable within conventional mechanics, such as the single-particle, energy-conserving, internal-redistribution relaxation mechanism discussed in detail in the paper.

We believe that our proposal for a resolution of the irreversibility dilemma is logically coherent and consistent with all the successful results of quantum mechanics, it provides a new perspective to the description of nonequilibrium phenomena, and it is definite and explicit enough to imply new detailed predictions that, at least in principle, should be experimentally verifiable.

APPENDIX A: FROM EQUATION (1) TO EQUATION (4)

In terms of the representation of state operators given by relation (2a), the eigenvalues of ρ are $p_+ = (1+r)/2$ and $p_- = (1-r)/2$ where $r = |\mathbf{r}| = p_+ - p_-$ and $r \le 1$. If $F(x)$ is a function of the real variable x, then operator $F(\rho)$ may be written as

$$F(\rho) = \frac{1}{r}[p_+ F(p_-) - p_- F(p_+)]I + \frac{1}{r}[F(p_+) - F(p_-)]\rho \qquad \text{(A1a)}$$

$$= \frac{1}{2}[F(p_+) + F(p_-)]I + \frac{1}{r}[F(p_+) - F(p_-)]\mathbf{r}\cdot\mathbf{R} \qquad \text{(A1b)}$$

and, in particular,

$$\rho \ln \rho = \frac{1}{4}\left(\ln\frac{1-r^2}{4} + r\ln\frac{1+r}{1-r}\right)I + \frac{1}{2}\left(\ln\frac{1-r^2}{4} + \frac{1}{r}\ln\frac{1+r}{1-r}\right)\mathbf{r}\cdot\mathbf{R} \qquad \text{(A2)}$$

$$\mathrm{Tr}\,\rho\ln\rho = \frac{1}{2}\left(\ln\frac{1-r^2}{4} + r\ln\frac{1+r}{1-r}\right) \qquad \text{(A3)}$$

Using relation (3a) and the commutation rules obeyed by the spin operators, we find

$$\tfrac{1}{2}\{H, \rho\} = \tfrac{1}{2}\hbar\Omega_0[\tfrac{1}{2}(\mathbf{\Lambda}\cdot\mathbf{r})I + \mathbf{\Lambda}\cdot\mathbf{R}] \qquad \text{(A4)}$$

$$\mathrm{Tr}\,\rho H = \tfrac{1}{2}\hbar\Omega_0\mathbf{\Lambda}\cdot\mathbf{r} \qquad \text{(A5)}$$

$$\mathrm{Tr}\,\rho H^2 = (\tfrac{1}{2}\hbar\Omega_0)^2 \qquad \text{(A6)}$$

$$\mathrm{Tr}\,\rho H\ln\rho = \tfrac{1}{4}\hbar\Omega_0\left[\ln\frac{1-r^2}{4} + \frac{1}{r}\ln\frac{1+r}{1-r}\right]\mathbf{\Lambda}\cdot\mathbf{r} \qquad \text{(A7)}$$

$$i[H, \rho] = -\hbar\Omega_0(\mathbf{\Lambda}\times\mathbf{r})\cdot\mathbf{R} \qquad \text{(A8)}$$

The right-hand side of equation (1a) becomes

$$-\frac{1}{\tau}\frac{1-r^2}{2r}\ln\frac{1+r}{1-r}\frac{\mathbf{r}\cdot\mathbf{R}-r_e\mathbf{\Lambda}\cdot\mathbf{R}}{1-r_e^2} \tag{A9}$$

Combining equation (1) with relations (A8) and (A9) and the fact that

$$\frac{d\rho}{dt}=\frac{d\mathbf{r}}{dt}\cdot\mathbf{R} \tag{A10}$$

Equation (4) follows immediately.

APPENDIX B: EXISTENCE AND UNIQUENESS OF SOLUTIONS

As shown in Appendix A, for ρ in the set of state operators ($\rho^\dagger=\rho\gtrsim 0, \operatorname{Tr}\rho=1$), equation (1) is equivalent to equation (4) that we rewrite here as

$$\frac{d\mathbf{r}}{dt}=\mathbf{F}(\mathbf{r})=\begin{cases}\Omega_0\mathbf{\Lambda}\times\mathbf{r}-\dfrac{1-r^2}{2r}\ln\dfrac{1+r}{1-r}\dfrac{\mathbf{r}-r_e\mathbf{\Lambda}}{1-r_e^2}, & \text{if } r<1\\ \Omega_0\mathbf{\Lambda}\times\mathbf{r}, & \text{if } r=1\end{cases} \tag{B1}$$

where $r=|\mathbf{r}|$, $r_e=\mathbf{\Lambda}\cdot\mathbf{r}$ and $\mathbf{r}\neq\pm\mathbf{\Lambda}$. We have seen that if $\mathbf{r}(t)$ is a solution of equation (B1) then r_e is a constant with $-1<r_e<1$ and equation (9) is satisfied, i.e.,

$$\frac{dr}{dt}=G(r)=\begin{cases}-\dfrac{1}{\tau}\dfrac{r^2-r_e^2}{1-r_e^2}\dfrac{1-r^2}{2r^2}\ln\dfrac{1+r}{1-r}, & \text{if } |r_e|\le r<1\\ 0, & \text{if } r=1\end{cases} \tag{B2}$$

Here we will show that, for each initial condition in the Bloch sphere, Equation (B1) admits of a unique solution defined for $-\infty<t<+\infty$ and lying entirely in the Bloch sphere.

In the open region $D=\{\mathbf{r}|r<1\}$, the functions $F_j(r_1, r_2, r_3)$ [$\mathbf{F}=(F_1, F_2, F_3)$, equation (B1)] and the partial derivatives $\partial F_j/\partial r_k$ are defined and continuous. Therefore, for each $\mathbf{r}_0$ with $r_0<1$ there exists a unique complete solution $\mathbf{r}(t)$ of equation (B1), defined for $a<t<b$, such that $\mathbf{r}(0)=\mathbf{r}_0$.

Because $G(r)<0$ in D, equation (B2) implies that $r(t)<r_0<1$ for every $0\le t<b$. Thus, the complete solution remains internal to D and bounded for every $0\le t<b$, and it must therefore extend to $b=+\infty$.

The solution remains on the plane orthogonal to $\mathbf{\Lambda}$ defined by the constant $r_e=\mathbf{\Lambda}\cdot\mathbf{r}$ and spirals toward the equilibrium point $\mathbf{r}_e=r_e\mathbf{\Lambda}$ without ever reaching it at any finite time. This can be seen, for example, by integrating Equation (B2) to obtain

$$t_e = \int_0^{t_e} dt = \lim_{r \to r_e} \int_{r_0}^{r} \frac{dr'}{G(r')} = +\infty \tag{B3}$$

because in r_e the function $1/G(r)$ is infinite of the same order as $1/(r_e - r)$.

The complete solution, defined for $a < t < +\infty$, either extends to $a = -\infty$ or else it approaches the boundary $FD = \{\mathbf{r} | r = 1\}$ of domain D as t tends to some finite $a < 0$. But the latter case is impossible because integration of equation (B2) yields

$$a = \int_0^{a} dt = \lim_{r \to 1} \int_{r_0}^{r} \frac{dr'}{G(r')} = -\infty \tag{B4}$$

where $r_0 < 1$. Relation (B4) may be verified by observing that in $r = 1$ the function $-1/G(r)$ is infinite of the same order as the function $1/g(r)$ where

$$g(r) = \frac{1 - r^2}{2} \ln \frac{1 + r}{1 - r} \tag{B5}$$

and

$$\lim_{r \to 1} \int_{r_0}^{r} \frac{dr'}{g(r')} = \lim_{r \to 1} \int_{r_0}^{r} d \ln \ln \frac{1 + r'}{1 - r'} = +\infty \tag{B6}$$

We conclude that for each $\mathbf{r}_0$ in D equation (B1) admits of a unique solution entirely contained in D for every $-\infty < t < +\infty$. As t tends to $+\infty$, the solution approaches a stable equilibrium point. As t tends to $-\infty$, the solution approaches the boundary FD. On the boundary FD of its domain of definition, equation (B1) admits of periodic solutions, the conventional quantum mechanical solutions, which precess around Λ at the angular frequency Ω_0 and lie entirely on FD. These periodic boundary solutions could fail to be unique only by leaving the boundary FD in finite time. But this is impossible because of relation (B4). It is also readily seen that, except for the fixed points $\mathbf{r} = \pm\Lambda$, all boundary solutions are unstable.

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