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Anomalous μ^+ Precession in Silicon[*]

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ABSTRACT

We have studied precession of polarized positive muons in quartz and silicon in transverse magnetic fields, via the asymmetric decay. We observed free muon precession and two-frequency muonium precession, as well as two anomalous precession frequencies apparent only in silicon.

Positive muons stopping in condensed matter virtually always capture electrons from the medium to form muonium ($\mu^+ e^-$) atoms.[1] In an external magnetic field, this spin 1/2 - spin 1/2 system has energy eigenvalues given by a Breit-Rabi diagram (Fig. 1). The four indicated transitions (ν_{12}, ν_{23}, ν_{14}, ν_{34}) manifest themselves as frequencies of muonium precession when the muons are initially polarized transverse to the external field. This precession can be monitored by detecting positrons from the muon decay ($\mu^+ \rightarrow e^+ \nu_e \bar{\nu}_\mu$) with a counter telescope in the plane of precession. Because the decay positrons are emitted preferentially along the muon spin, the probability of detecting a positron will include a contribution which oscillates at the precession frequency. Repeated measurements of the time between muon stop and positron emission generate a histogram whose Fourier transform exhibits peaks corresponding to the above-mentioned precession frequencies.

Because of the limited time resolution of the experimental apparatus (~1 nsec), only ν_{12} and ν_{23} (the two lowest of the four allowed transitions) can be observed, and these only for sufficiently low magnetic fields. Muonium was first observed in this way by Gurevich et al.[2] in quartz and cold (77°K) germanium.

The muonium hyperfine frequency ν_0 can be extracted from a measurement of the two observable precession frequencies for muonium. In vacuum, $\nu_0 = 4463$ MHz.[3] Gurevich et al. found that ν_0 (quartz) $= \nu_0$ (vac), but that $\nu_0(\mathrm{Ge})/\nu_0(\mathrm{vac}) = 0.56 \pm 0.01$. We have verified their result for muonium in quartz and measured ν_0 in silicon at 77°K. We find that $\nu_0(\mathrm{Si})/\nu_0(\mathrm{vac}) = 0.45 \pm 0.02$, in agreement with Andrianov et al.[4] The Ge and Si results have been interpreted by Wang and Kittel[5] in terms of a swelling of the interstitial muonium atom due to shielding by valence band electrons. Identification of interstitial ground state muonium as a deep donor clarifies the nature of hydrogen-like impurities in these materials.

In our cold p-type silicon spectra, we see not only the two familiar muonium peaks, but also two others of similar amplitude, which we have called "anomalous muon precession" for lack of a positive identification of their source. Figure 2 shows a comparison between Fourier spectra for silicon and fused quartz in the same field, demonstrating the absence of anomalous precession in quartz. Whereas the muonium frequencies rise approximately linearly with field up to a few hundred gauss, and are independent of the orientation of the crystal in the field, the anomalous frequencies have the field dependence shown in Fig. 3, and are slightly anisotropic, as indicated. Both anomalous precession and muonium precession have a lifetime on the order of 300 nsec. Neither of these signals has been detected in n-type Si at 77° K or in any silicon sample at room temperature.

The anomalous frequencies are much higher than the free muon precession frequencies in weak magnetic fields. The muon must therefore be coupled to a particle or system with a larger magnetic moment than its own, as in muonium, where it is coupled to an electron by the contact interaction. The field dependence of the data can in fact be fitted to frequencies ν_{12} and ν_{34} of a modified Breit-Rabi formula (see Fig. 1), if the different crystal orientations are treated as separate cases. However, it is necessary to allow both the hyperfine coupling strength and the g-factor of the electron to vary in order to obtain a fit. For the case of the [111] crystal axis parallel to the field, the best value for $\nu_0/\nu_0(\text{vac})$ is 0.0198 ± 0.0002; for [100] parallel to the field, the best value is $\nu_0/\nu_0(\text{vac}) = 0.0205 \pm 0.0003$. In both cases the best value for g_e is 13 ± 3. Clearly, the spin g-factor of an electron cannot be much different from 2, nor can a pure contact interaction be anisotropic; this modified Breit-Rabi description is meant only as a phenomenological characterization of the data.

These results can be interpreted in terms of a number of physical models. Perhaps the simplest is shallow-donor muonium. Here the electron wave function is spread over many lattice sites, whereas the entire deep-donor muonium atom fits into one interstitial site. An s-state cannot produce the observed behavior, due to the relatively invariable spin g-factor of the electron. However, in the 2p state the orbital g-factor can be large and anisotropic: the electron wave function for a shallow donor must be a superposition of conduction band states, which may have small anisotropic effective masses. If the spin-orbit coupling for the electron is large, j_e becomes a good quantum number, and $\underset{\sim}{J}^e$ formally replaces $\underset{\sim}{S}^e$ in the Breit-Rabi Hamiltonian. A possible objection to this model is the requirement of a minimum lifetime of ~300 nsec for the 2p excited state. Hindrance of the normally fast radiative E1 transition 2p → 1s can be explained by the small overlap between electron wave functions in the shallow-donor 2p state and the deep-donor 1s state.

A second physical model is suggested by the large variety of ESR centers which have been observed in radiation-damaged silicon.[6] The muon may create a paramagnetic lattice defect (e.g., a broken bond) at the end of its range, combining with it to form a muon-defect bound state. Such a center can also be described by a modified Breit-Rabi Hamiltonian.

The possibility that the anomalous precession is due to formation of a bound state of a muon with an impurity atom is considered remote. The fractional concentration of impurity atoms in our sample is $\sim 10^{-8}$ or less; muons can be expected to slow from ~100 eV to thermal velocities within $\sim 10^3$ collisions.[7] Thus the probability of a muon passing within several lattice sites of an impurity atom at subionizing velocity is negligible. Furthermore, the time for deep-donor muonium atoms to diffuse to impurity atoms with muon affinities must be longer than ~300 nsec, the observed relaxation time for muonium precession.

However, in stopping, the muon must generate a high density of free electrons and holes, with which it may subsequently combine. If we regard the μ^+ as a positive impurity ion in an interstitial position, observations of impurity-exciton bound states in silicon[8] provide a precedent for two models involving excitons. The first model is the neutral muonium-exciton molecule

($\mu^+ e^- e^- h^+$), in which the two electrons are assumed to have paired spins, in analogy with ground-state H_2. The μ^+ is thus coupled to the hole by a dipole-dipole interaction. Orientational effects are predicted by this model if the molecule is "pinned" by being wedged into an oblong interstitial site in the unit cell.[9] A second model of this type is the ionized muonium-exciton molecule ($\mu^+ e^- h^+$), in which all three particles are coupled via contact interactions. These models draw support from the fact that measured free exciton lifetimes in silicon at 80°K are about 400 nsec.[10]

None of the above physical models for anomalous muon precession can be eliminated on the basis of existing data; however, we feel that shallow-donor 2p muonium is the most probable explanation. In an earlier study at Columbia,[11] the "quenching" of μ^+ depolarization in silicon by a magnetic field applied parallel to the muon polarization was interpreted in terms of transitory muonium formation. Their results in p-type silicon at ≤ 77°K suggested the existence of two species of muonium with different hyperfine couplings. However, their prediction that muonium in silicon would only form a short-lived shallow-donor state is contradicted by our observation of long-lived deep-donor 1s muonium. If the anomalous precession is in fact due to shallow-donor 2p muonium (albeit long-lived), their conclusions will be at least partially vindicated. In any event, it is clear that positive muons can provide a great deal of new information about the behavior of hydrogen-like impurities in silicon.

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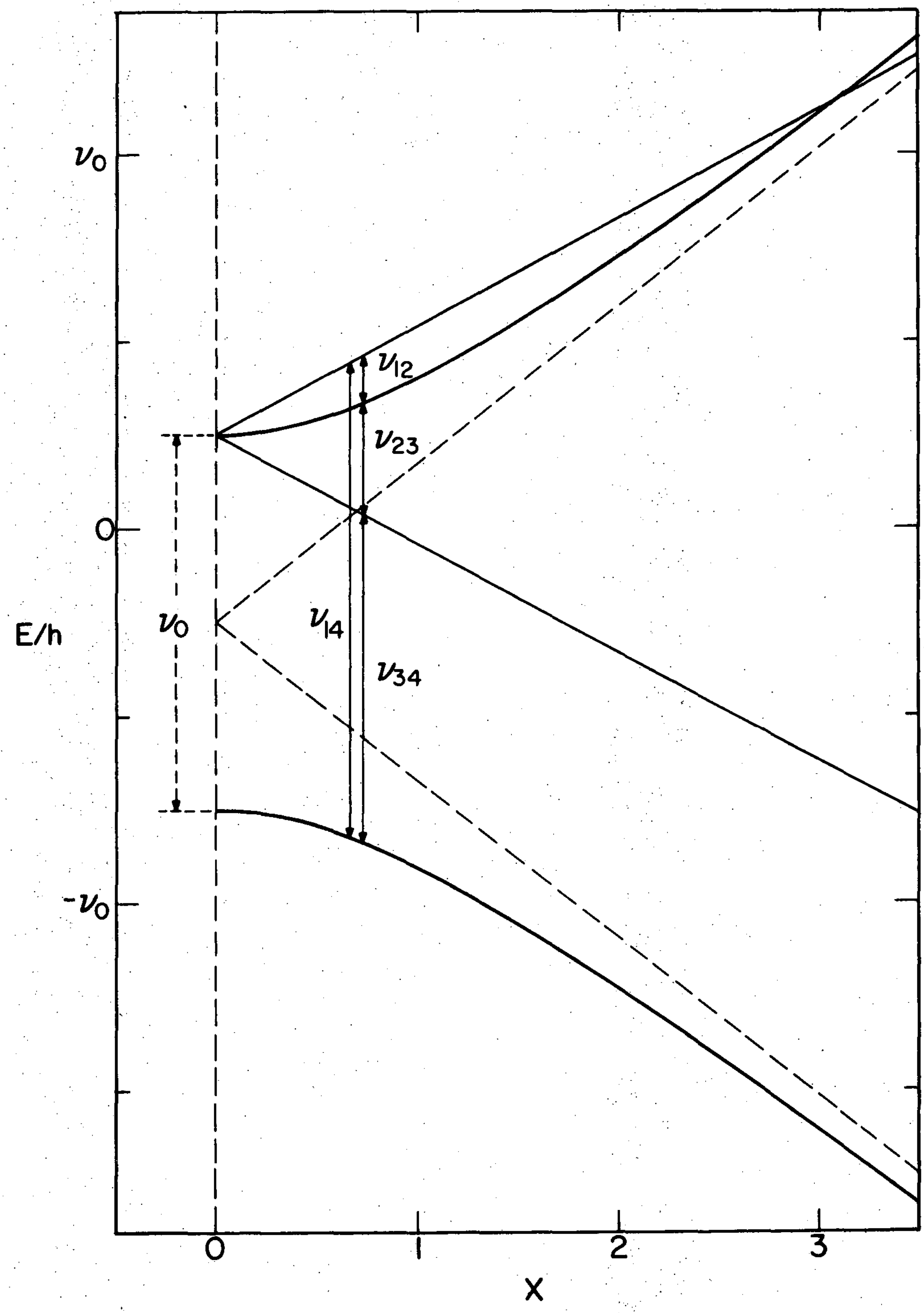
FIGURE CAPTIONS

FIG. 1. Energy eigenstates of $\ell = 0$ muonium in an external magnetic field, as functions of the dimensionless "specific field" $X = 2\mu_e B/h\nu_0$. For graphical clarity, an unphysical value of m_μ/m_e is used to generate the plot. The four allowed transitions are indicated. The relation $\nu_{12} + \nu_{34} = \nu_0$ holds for all fields, where ν_{12} is understood to be negative for $X > X^* \approx (m_\mu - m_e)/2m_e$, where the top two levels cross.

FIG. 2. Frequency spectra of muons in fused quartz at room temperature and in p-type silicon at 77° K. In both cases the applied field is 100 G. The vertical axis is the square of the Fourier amplitude, in arbitrary but consistent units. In the lower graph the vertical scale is expanded by a factor of 10 to the right of the dashed line. The prominent peaks (from left to right) are: the free muon precession signal at 1.36 MHz; a characteristic background signal at 19.2 MHz, due to rf structure in the cyclotron beam; the two anomalous frequencies at 43.6 ± 2.9 MHz (silicon only); and the two 1s muonium peaks centered about 139 MHz. The wider splitting of the two 1s muonium lines in silicon is due to the weaker hyperfine coupling. These spectra were produced by Fourier-analyzing the first 750 nsec of the experimental histograms. For comparison, the muon asymmetries obtained by maximum-likelihood fits to the first 5 μsec of data were 3.81% ± 0.35% for quartz and 5.05% ± 0.63% for p-type Si at 77° K.

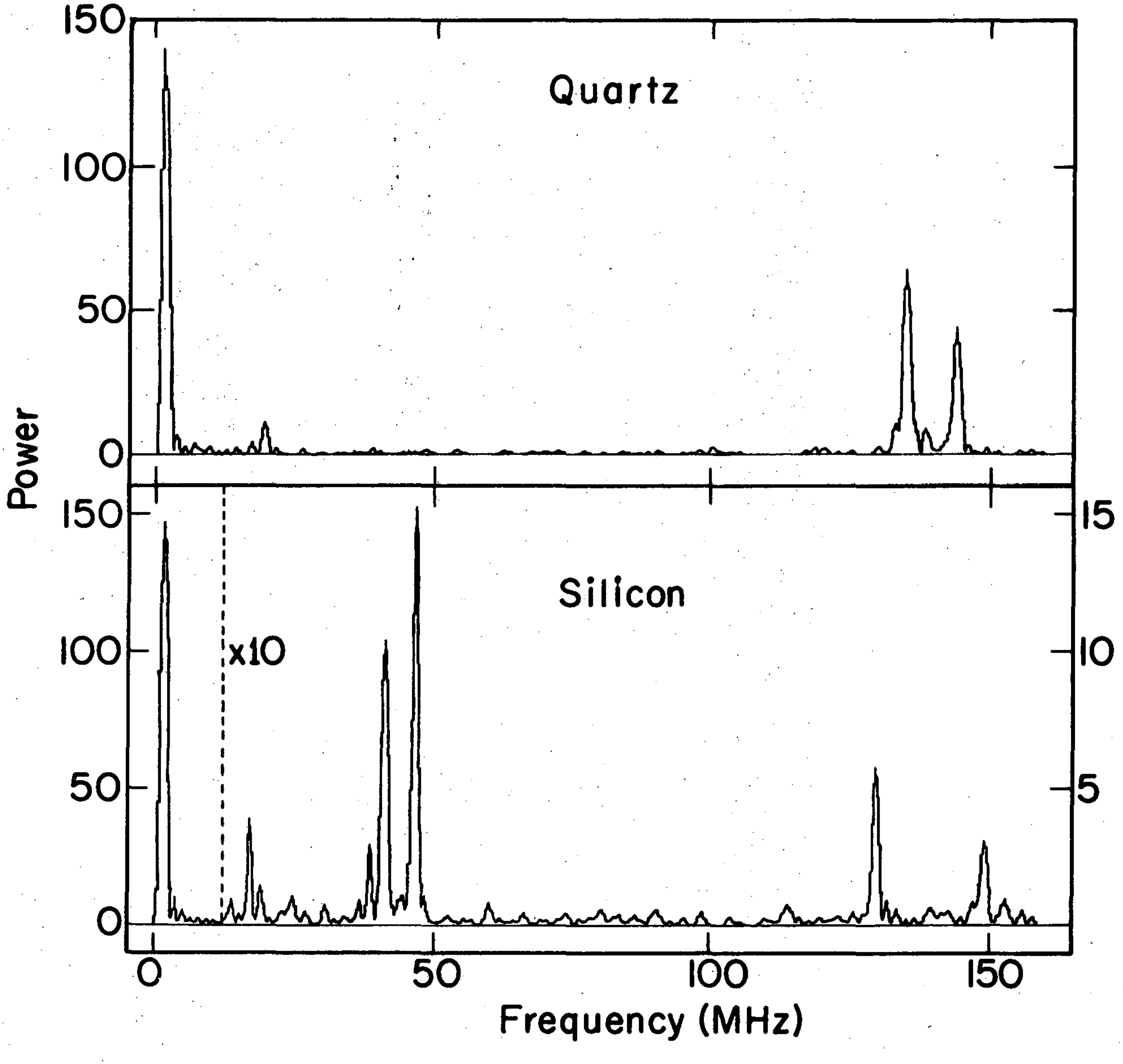
FIG. 3. Dependence of anomalous frequencies in silicon upon field strength and crystal orientation. Round points and solid lines are data and best fit for [111] crystal axis along the field; triangular

points and dashed lines are data and best fit for [100] axis along the field. Free muon, 1s muonium, and cyclotron background signals are not shown. A number of peaks appear in the spectra in addition to the fitted "proper" anomalous frequencies; these are unexplained. They are indicated by square points (for prominent peaks) and horizontal bars (for weak or questionable peaks). The higher of the "proper" anomalous frequencies is missing at several fields. This is because the spectra showed no statistically significant peaks at those positions.



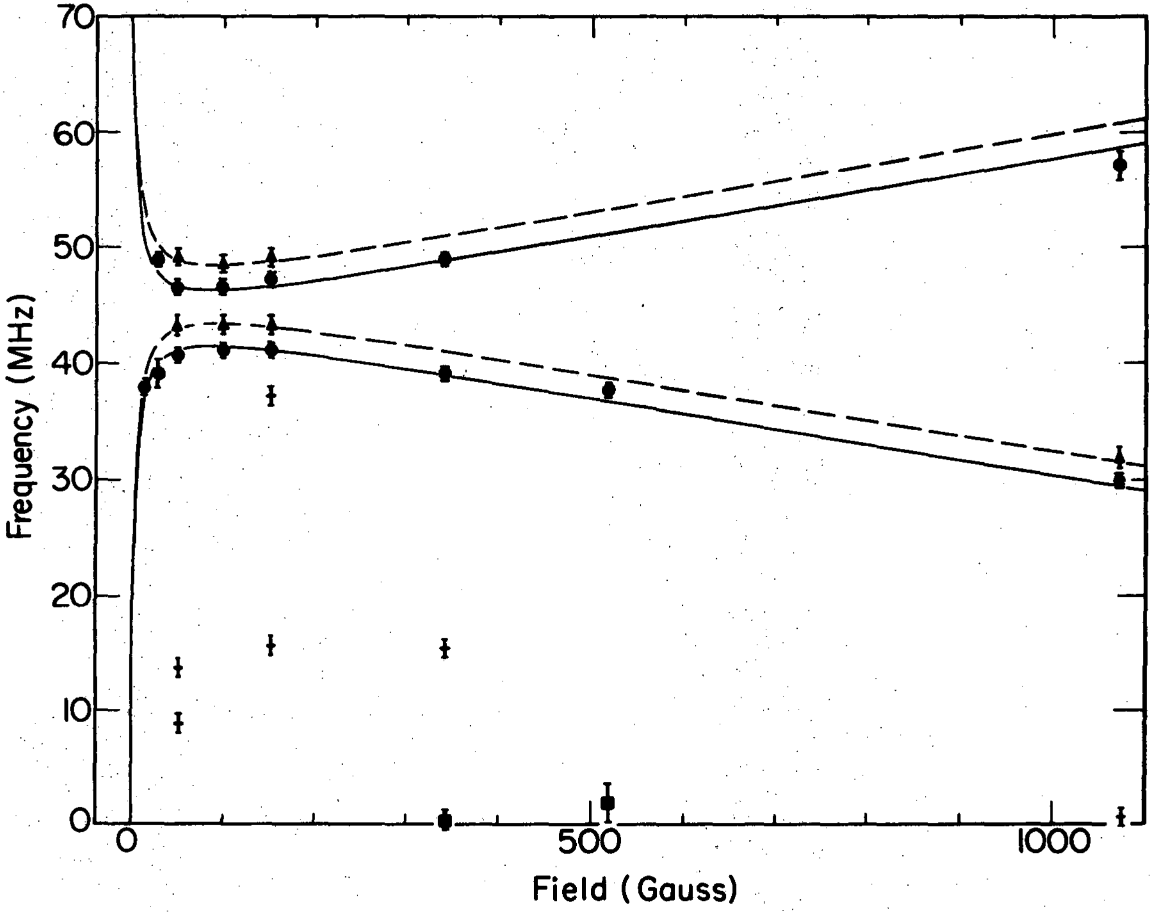
XBL733-2502

Fig. 1



XBL734-2593

Fig. 2



XBL 734-2592

Fig. 3

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