Decoupling of Muonium in High Transverse Magnetic Fields

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Depolarization of muonium due to superhyperfine interaction, which until now has severely impeded the study of muonium in solids, can be quenched in a high transverse magnetic field (1.2 T). We report the observation of muonium precession in single crystals of KCl, CaF_2, GaP, and Al_2O_3. The high-field frequencies are of order 2 GHz and provide a direct and accurate (60 ppm) measure of the hyperfine interval. Several novel features of the muonium centers are observed.

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Muonium (μ^+e^-, Mu), a light hydrogenlike atom, is studied by the technique of muon spin rotation (μSR).[1] The Mu hyperfine frequency, ν_0, is a measure of the electron spin density at the muon and may be considerably different in a solid compared with vacuum. Precise measurements of ν_0 are necessary to (1) distinguish between different Mu states in the lattice, (2) measure isotope shifts relative to hydrogen, and (3) test calculations of the electronic structure of the simplest atomic impurity. In addition, important information on the Mu site and its mobility can be obtained from the Mu depolarization rate.

Until now almost all such information on Mu centers in solids has been derived from the precession frequencies in low magnetic fields (ν_{12} and ν_{23} in Fig. 1) and more recently from the zero-field oscillations[2,3] (ν_{24} in Fig. 1). However, these frequencies have been observed in only a few oxide insulators,[1,4] solid noble gases,[5] and elemental semiconductors.[6] In the vast majority of solids that contain an abundance of nuclei with magnetic moments, these signals are unobservable because of the superhyperfine (SHF) interaction with surrounding nuclear spins. The SHF interaction causes splittings in the hyperfine levels which in low fields, where the muon and electron spins are strongly coupled, lead to a large number of small-amplitude frequencies that are not resolved. In such cases a limited amount of information on the hyperfine coupling and SHF coupling of the Mu center has

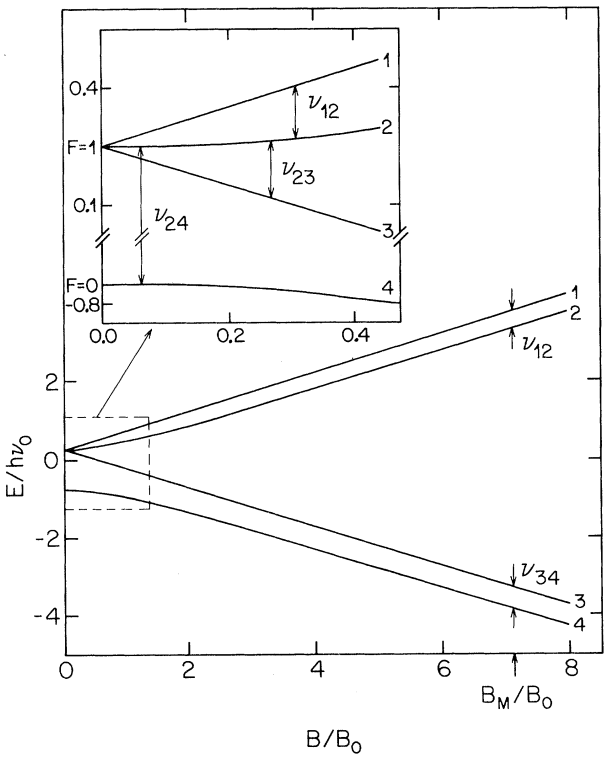

FIG. 1. Breit-Rabi diagram for Mu. Energy is in units of $h\nu_0$ (ν_0=4463.302 MHz in vacuum) and magnetic field in units of B_0 (0.1585 T in vacuum). The high-field frequencies ν_{12} and ν_{34} in the Paschen-Back region ($B/B_0 >> 1$) have a broad maximum and minimum, respectively, at B_M.

been obtained through decoupling experiments in a longitudinal magnetic field,[7,8] applied parallel to the initial muon polarization. However, only the magnitude of the total muon polarization is observed with this latter technique whereas detailed investigations require frequency measurements that distinguish between various states.

We report here the observation of muonium precession in single crystals of KCl, CaF_2, Al_2O_3, and GaP despite the presence of nuclear moments. Most of the measurements were performed in a magnetic field of 1.2 T in the Paschen-Back region where the Mu electron spin is decoupled from the muon and neighboring nuclear spins. In this field regime the frequency spectrum reduces to the simple one observed in the absence of a SHF interaction, consisting of only two frequencies with equal precession amplitude (ν_{12} and ν_{34} in Fig. 1):

$$\nu_{12}=\tfrac{1}{2}\nu_0+\Gamma_- B-\tfrac{1}{2}\nu_0[1+(B/B_0)^2]^{1/2},$$
$$\nu_{34}=\tfrac{1}{2}\nu_0-\Gamma_- B+\tfrac{1}{2}\nu_0[1+(B/B_0)^2]^{1/2}, \quad (1)$$

where $\Gamma_\pm=(|g_e|/m_e\pm|g_\mu|/m_\mu)e/8\pi$, B_0 (equal to $\nu_0/2\Gamma_+$) is the hyperfine field of the muon at the electron, and B is the applied magnetic field. Note that the hyperfine frequency ν_0 is equal to the sum of ν_{12} and ν_{34}. At the special field

$$B_M=[(\Gamma_+/\Gamma_-)^2-1]^{-1/2}\nu_0/2\Gamma_+ \quad (2)$$

(=1.13 T in vacuum) the Mu frequencies ($\nu_{12}=1.923$ GHz and $\nu_{34}=2.541$ GHz, in vacuum) have a broad maximum and minimum, respectively, as a function of magnetic field, and thus are insensitive to field inhomogeneity arising from the magnet or from local dipolar fields.

Spin-polarized muons of initial momentum 80–105 MeV/c were stopped in the samples. A small iron-core electromagnet (24 mm gap and 80 mm pole face diameter) was specifically designed to limit stray fields to less than 0.01 mT at a distance of 100 mm from the magnet center. This permitted the use of short light guides between the fast plastic scintillators and photomultipliers, an essential feature in achieving the timing resolution necessary to observe such high frequencies.

A Fourier power spectrum of the μSR time histogram in single-crystal KCl (in which all nuclei have spin $\frac{3}{2}$) is shown in Fig. 2. Both high-field Mu frequencies ν_{12} and ν_{34} are clearly visible, as well as the bare muon frequency at $\nu_\mu=152.5$ MHz. The reduction in amplitude of ν_{34} relative to ν_{12} is consistent with a Gaussian timing resolution function[3] of 230 ps full width at half maximum. The reduced hyperfine interval ν_r at 291 K (see Table I) is 650 times more accurate than the previous longitudinal field measurement[8] of 0.97(4) and thus allows a meaningful comparison with EPR measurements of hydrogen (H). Two paramagnetic H centers have been observed in pure KCl: the interstitial or U_2 center[9] ($\nu_r=0.9704$ at 77 K) and the Cl-substituted

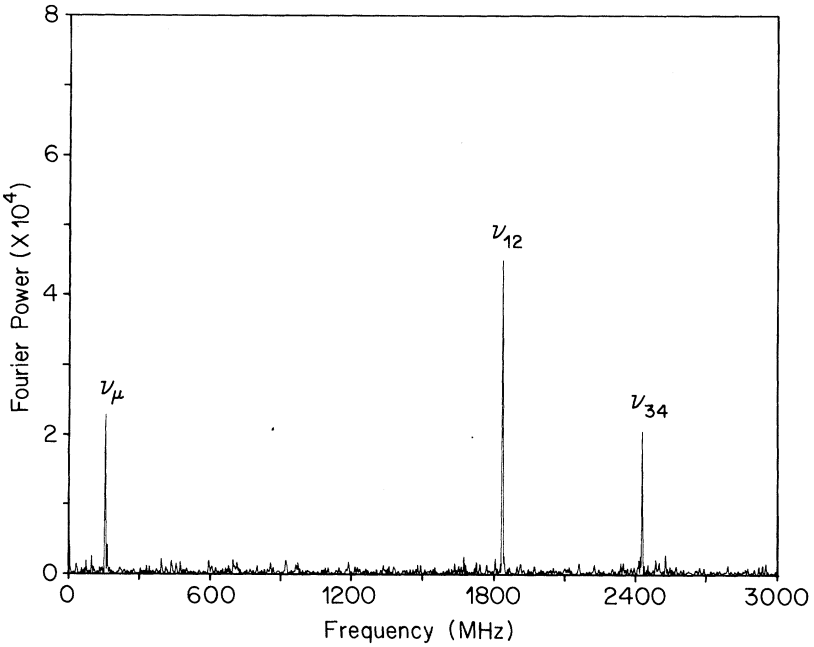

FIG. 2. Fourier power spectrum in KCl in a 1.12-T transverse magnetic field at 300 K. The two frequencies ν_{12} and ν_{34} are due to Mu whereas the frequency ν_μ is due to muons in a diamagnetic environment.

TABLE I. The reduced hyperfine interval[a] (ν_r), muonium fraction[b] (F_{Mu}), and depolarization rate (λ) measured in high transverse fields.

Sample	Temp. (K)	Field (T)	ν_r[a]	F_{Mu}[b] (%)	λ (μs^{-1})
KCl	14	1.20	0.959 05(8)	23(3)	0.3(4)
KCl	291	1.12	0.954 34(6)	69(5)	2.1(5)
CaF_2	10	1.13	1.003 6(10)	64(14)	49(12)
CaF_2	300	1.13	1.016 83(22)	73(17)	16(2)
Al_2O_3	10	1.13	0.988 95(51)	10(2)	4(3)
GaP	10	1.20	0.652 9(11)	72(10)	100(20)
GaP	140	1.20	0.619 7(5)	54(5)	58(4)

[a]The hyperfine interval divided by the vacuum value (4463.302 MHz for Mu).

[b]Total precession amplitude corrected for finite timing resolution and normalized to the free muon precession amplitude in Cu.

or U_3 center[10] ($\nu_r = 0.9916$ at 20 K). Comparing our interpolated results at these temperatures we find a shift in ν_r of -1.20% relative to the U_2 center or -3.26% relative to the U_3 center. We attribute this shift to the larger zero-point motion of Mu which has one-ninth the mass of H. A similar isotope shift of -1.3% for Mu relative to H has been observed in SiO_2.[3]

The high-field frequencies can be used to measure small changes in the hyperfine interval as a function of the temperature. As an example, the temperature dependence of the hyperfine interval in KCl is shown in Fig. 3. The data were fitted by a Debye model of the Mu-phonon interaction,[2,11,12]

$$\nu_0(T) = \nu_0(0)\left[1 - C\left(\frac{T}{\theta_D}\right)^4 \int_0^{\theta_D/T} \frac{x^3}{e^x - 1}\,dx\right], \tag{3}$$

yielding $\nu_0(0) = 4280.6(2)$ MHz, $C = 0.0159(15)$, and an effective Debye temperature $\theta_D = 229(17)$ K which is in agreement with the Debye temperature of 236 K derived from specific-heat data.[13] This indicates that a single Mu center is being observed at all temperatures and that interaction with the long-wavelength acoustic phonons is responsible for the temperature dependence in ν_0.

The SHF interaction itself can be used to obtain information on the site of Mu and its mobility in the lattice. This is accomplished by reducing the applied field until the muon and electron are sufficiently coupled to produce a measurable depolarization due to SHF interaction. For example the depolarization rate, λ, was measured in KCl as a function of temperature at 1.12 T and at 0.159 T (see Fig. 3). At 1.12 T, depolarization due to SHF interaction is completely quenched at all temperatures. The slight increase in λ at 291 K was observed to be independent of magnetic field down to 0.3 T and therefore must be due to some additional depolarization mechanism such as trapping at impurities. At the lower field of 0.159 T, the depolarization rate is considerably larger and increases as the temperature is decreased (see Fig. 3). The larger depolarization rate is attributed to the SHF interaction which now causes considerable line broadening since the muon and the electron are not

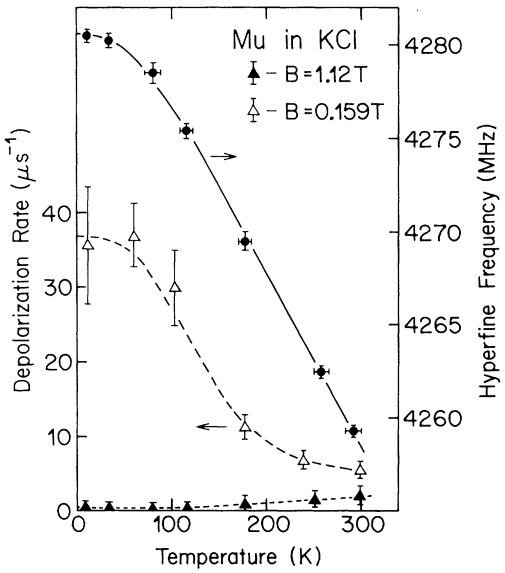

FIG. 3. Temperature dependence of the hyperfine frequency at 1.12 T (circles) and the depolarization rate at 1.12 T (closed triangles) and at 0.159 T (open triangles). The solid curve is a fit by Eq. (3). The dashed curves are guides to the eye.

completely decoupled. The complete field dependence of λ at 291 K (not shown here) indicates an effective isotropic SHF interaction coupling of 3.3(4) MHz compared with 23.6 MHz for interstitial hydrogen.[9] However, it is clear from the increase in λ at low temperatures in an applied field of 0.159 T that the strength of the effective SHF interaction increases as the temperature is decreased. This suggests that the Mu frequency lines are motionally narrowed at the higher temperatures leading to an effective SHF interaction coupling smaller than for the static case. This implies that the observed center is a rapidly diffusing interstitial, i.e., the muonic analog of the U_2 center.

An increase in the polarized Mu fraction by more than a factor of 2 was observed in KCl between 14 and 291 K (see Table I). A model in which the Mu formation rate is comparable to the precession frequencies and increases with temperature explains this unusual behavior. A complete description of the data and model will be presented later. Although such Mu formation after thermalization has been postulated[14] this is the most direct evidence of such a process.

The small depolarization rate in KCl at 291 K in high field (see Table I) is at variance with the interpretation of longitudinal field measurements[8] in terms of the Ivanter-Smilga theory,[15] according to which the Mu reacts chemically and undergoes spin exchange at rates in excess of 1000 μs^{-1}. Our observation of a long-lived coherent Mu state places an upper limit of about 3 μs^{-1} on the rate for such processes.

We have also observed high-field Mu precession in single crystals of CaF_2, GaP, and Al_2O_3 for which no Mu precession has been reported in low fields. The results are summarized in Table I. Detailed investigations will be reported later, but there are several novel features that we wish to point out here:

(1) The hyperfine interval of Mu in CaF_2 at 10 K is 1.32% lower than at 300 K. Remarkably, this temperature shift is opposite to that of interstitial H in CaF_2 for which the hyperfine interval at 77 K is 0.5% higher than at 300 K.[12]

(2) The hyperfine interval of Mu in Al_2O_3 at 10 K is consistent with that indicated by the longitudinal field measurements[7] at 300 K [$\nu_r = 1.04(7)$]. At 10 K only 10(2)% of the muons form Mu while 76(5)% are in a bare muon state. However, at room temperature neither Mu nor μ^+ precession was observed.

(3) In the compound semiconductor GaP the small ν_r is consistent with results in the elemental covalent semiconductors.[6] Note the enormous temperature shift of -5% in the hyperfine interval between 10 and 140 K compared to KCl where the shift is only 0.19% over this temperature range.

In conclusion, we have demonstrated a powerful high-transverse-field technique which provides detailed information of Mu states in condensed matter. Spin depolarization due to SHF interaction with nuclear spins, which severely limits the low-field technique, is quenched in a high transverse field. A large body of new information on Mu in insulators and semiconductors is now accessible.

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[1]J. H. Brewer and K. M. Crowe, Annu. Rev. Nucl. Sci. **28**, 239 (1978).

[2]E. Holzschuh, Phys. Rev. B **27**, 102 (1983).

[3]E. Holzschuh, W. Kündig, and B. D. Patterson, Helv. Phys. Acta **54**, 552 (1981).

[4]D. P. Spencer, D. G. Fleming, and J. H. Brewer, in Proceedings of the Yamada Conference on Muon Spin Rotation (to be published).

[5]R. F. Kiefl, J. B. Warren, G. M. Marshall, and C. J. Oram, J. Chem. Phys. **74**, 308 (1981).

[6]E. Holzschuh, W. Kündig, P. F. Meier, B. D. Patterson, J. P. F. Sellschop, M. C. Stemmet, and H. Appel, Phys. Rev. A **25**, 1272 (1982).

[7]E. V. Minaichev *et al.*, Zh. Eksp. Teor. Fiz. **58**, 1586 (1970) [Sov. Phys. JETP **31**, 849 (1970)].

[8]I. G. Ivanter *et al.*, Zh. Eksp. Teor. Fiz. **62**, 14 (1972) [Sov. Phys. JETP **35**, 9 (1972)].

[9]J. M. Spaeth, Z. Phys. **192**, 107 (1966).

[10]W. Hayes and J. W. Hodby, Proc. Roy Soc. London, Ser. A **294**, 359 (1966).

[11]E. Simanek and R. Orbach, Phys. Rev. **145**, 191 (1966).

[12]S. G. Sligar and H. Blum, Phys. Rev. B **3**, 3587 (1971).

[13]J. T. Lewis, Phys. Rev. **161**, 877 (1967).

[14]P. W. Percival, Hyperfine Interact. **8**, 315 (1981).

[15]I. G. Ivanter and V. P. Smilga, Zh. Eksp. Teor. Fiz. **54**, 559 (1968) [Sov. Phys. JETP **27**, 301 (1968)].