

sumed direct 1:1 correspondence between them and fractional compositions, leading to sizable overestimates of Lorentzian parentage.

¹⁶E. J. McGuire, *Phys. Rev. A* **3**, 1801 (1971).

¹⁷Wertheim, Butler, West, and Buchanan, Ref. 15.

¹⁸W. Meyer, *J. Chem. Phys.* **58**, 1017 (1973).

¹⁹U. Gelius, E. Basilier, S. Svensson, T. Bergmark,

and K. Siegbahn, *J. Electron Spectrosc. Relat. Phenomena* **2**, 405 (1974).

²⁰The S Franck-Condon overlap factors can be shown to obey a Poisson distribution (Ref. 1), the standard deviation of which is \sqrt{S} ; the full linewidth at half-maximum of such a distribution is proportional to $\hbar\omega_{LO}\sqrt{S} = (\Delta E\hbar\omega_{LO})^{1/2}$, the leading term in Eq. (1).

Probing the Superconducting Vortex Structure by Polarized- μ^+ Spin Precession

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The magnetic field structure of vortices in the mixed state of a lead-indium alloy and of niobium was probed by stopped μ^+ particles. Fourier transforms of time-dependent spin-precession data contain a peak at the average internal field, which is also the applied field. For the alloy only there is a second peak at the saddle point of the internal field distribution. Motional averaging due to quantum diffusion of the muons at low temperatures can account for these results.

Recent experiments have demonstrated the usefulness of using polarized muons as microscopic probes of local magnetic fields in magnetic metals.¹ Positive muons can be considered as radioactive protons with known magnetic moment ($e\hbar/2m_\mu c$) and lifetime (2.2 μsec). The field on the muon is easily determined from the precession frequency of the anisotropic angular distribution of decay positrons ($\mu^+ \rightarrow e^+ + \nu_e + \bar{\nu}_\mu$). By analogy to H^+ , the μ^+ is presumed to reside at interstitial sites in metals.²

The muon spin-precession technique is analogous to the well-known time-dependent perturbed-angular-correlation method for common radioactive species. It was used to study the local magnetic field distribution in samples of $\text{Pb}_{0.90}\text{In}_{0.10}$ and Nb, type-II superconductors, in the mixed state. Our results indicate that the μ^+ particle remains highly mobile at low temperatures in Nb.

Previous experiments on the magnetic structure of the mixed state of type-II superconductors made use of such techniques as NMR,³ neutron diffraction,⁴ perturbed angular correlation,⁵ molecular beams,⁶ and decoration microscopy.⁷ From the results of such experiments, the prominent features of the mixed-state local field distribution expected in the muon-precession-frequency spectrum are a peak at the local field cor-

responding to the saddle-point location in the vortex lattice, B_s , and maximum and minimum cut-off fields.³ The periodic inhomogeneity created is expected to lead to both line broadening and a shift, since $B_s < B$, the average of the internal field. Experiments reported in this Letter were motivated by the attractiveness of applying a versatile new technique to a model example and the possibility of studying various dynamical effects.

Predictions of what the precession data should look like have been calculated before,⁸ but we have preferred a semiempirical approach. The precession angle as a function of time at a site \vec{r} in the vortex lattice can be derived from the expression

$$\theta(t) = \gamma_\mu t B \sum_{\vec{g}} F_{\vec{g}} \cos(\vec{g} \cdot \vec{r}),$$

where γ_μ is the gyromagnetic ratio of the muon and \vec{g} a reciprocal-vortex-lattice vector. Choosing zero time to correspond to zero phase is arbitrary. Values of the Fourier components $F_{\vec{g}}$ for our niobium specimen were interpolated from neutron-diffraction measurements on specimens of comparable purity.⁹ Similarly, measurements on an alloy of comparable Ginzburg-Landau parameter were used as a basis for calculating appropriate Fourier components for the lead-indium specimen.¹⁰

When $\cos\theta(t)$ is numerically averaged over the vortex unit cell and Fourier transformed, one calculates a frequency spectrum similar to the distribution function of local fields. In generating such transforms for various fields and temperatures, only the initial 4 μsec in time were used, since that is also the time interval over which our data extend. These calculations predict the amplitude and position of the expected peak at the saddle-point field. Possible influence of deformations of the vortex structure and normal depolarization effects were not included. Modifications by muon mobility are considered below.

The two specimens are 7.6-cm square plates which were cold rolled, annealed, and gold electroplated.¹¹ The specimens were mounted in a square copper frame oriented at a 30° angle to the applied magnetic field direction, in good thermal contact with a liquid-helium cryostat. The projected cross section of the aperture of the frame was 40 cm^2 , while the cross section of the detector immediately in front of the cryostat was 25 cm^2 .¹ Surrounding the sample were a copper radiation shield with two windows of 0.15-mm-thick copper foil and an aluminum vacuum can 1.6 mm thick. Cryostat parts presented a maximum 25% contribution to the total number of mu-

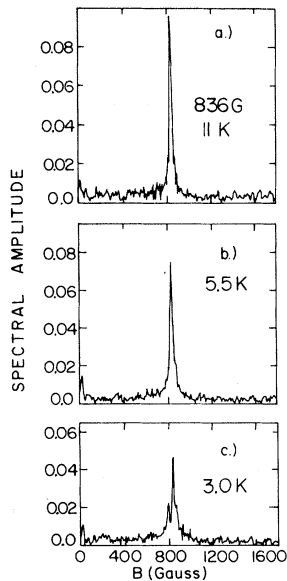


FIG. 1. Fourier-transform amplitudes for normalized reduced precession data, for the lead-indium alloy (a) in the normal state ($T_c = 7.0\text{ K}$), and (b), (c) field cooled into the mixed state. (b) and (c) have a factor of 2 better statistics than (a). The same is true for Fig. 2.

on stops in the apparatus. Since the diamagnetic moment is small for the geometry chosen, B is nearly equal to the applied field. The field is perpendicular to the plane of the muon precession.

Data were acquired according to one of the following three procedures: (1) applying the field in the normal state; (2) cooling in a steady field into the mixed state—referred to as “field cooling”; (3) cooling in zero field, followed by turning on the field—“zero-field cooling.” Several temperatures and fields below $H_{c2}(T)$, the temperature-dependent critical field, were used for each specimen.

The time-dependent decay spectra were first fitted in order to subtract a time-independent background term, divide out the 2.20- μsec muon-exponential-decay factor, and subtract the dc component. Then, after dividing the results by the coefficient of the exponential term, a fast Fourier analysis was done. The Fourier spectral amplitudes thus obtained are normalized to the number of events. These spectra as a function of the effective local field seen by the muon are shown in Figs. 1 and 2 for the lead-indium and niobium specimens, respectively. A precession asymmetry of approximately 0.20 was typically observed in the normal state. However, depolarization times on the order of 2.5 μsec resulted in a factor of 2 reduction in the normalized

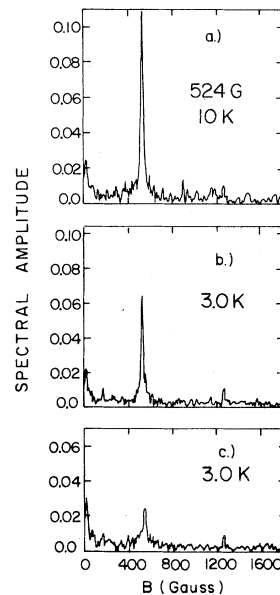


FIG. 2. Fourier transform spectra for niobium: (a) normal state ($T_c = 8.8\text{ K}$), (b) field cooled into the mixed state, and (c) zero-field cooled.

amplitude, in evidence in Figs. 1(a) and 2(a). Inhomogeneity in our applied field can account for this observation.

In Fig. 1 one observes that upon field cooling lead-indium at 836 Oe into the mixed state at 5.5 K, line broadening occurs. Upon further cooling to 3.0 K, a bifurcation is easily resolved. The satellite peak at 3.0 K, shifted by 45 G to lower field, is identified as originating from the saddle-point peak in the local-field distribution at B_s . (It is not resolved at 5.5 K.) The corresponding calculated shift is 58 G and the calculated magnitude of this peak exceeds the measured magnitude by 20%. General features of these and additional data taken at other fields and temperatures show that the strengths of the components at the applied field are somewhat larger than expected, i.e., 25% of the normal-state observation, while the saddle-point components are somewhat smaller. Observed positions of B_s are in good agreement with the calculations.

Quite different results were found in niobium, Fig. 2. First, no bifurcation was observed in any of the data taken below T_c . Secondly, the amplitude of the line observed in the mixed state, unshifted with respect to B , has a larger amplitude than the same line in the lead-indium spectra.

Several zero-field-cooled experiments, run on both specimens, show that our third procedure results in a single weak line, e.g., Fig. 2(c). We believe that the spectrum for the nonuniform vortex structure, prepared this way, is smeared out¹² and that the observed signal comes from stops in the cryostat.

Additional data for Nb taken at 524 Oe and 6.5 K and at 2000 Oe and 3.0 K gave results similar to those of Figs. 2(b) and 2(c). At the higher temperature or field, the saddle-point peak is expected to be larger, well above the noise, yet none appeared in any of the spectra.

Motional-narrowing effects of possible muon diffusion among the interstitial sites is known to potentially have a major impact upon the results.¹³ A consistent picture explaining our finding strong lines at the average field would incorporate rapid translational motion of the muon at low temperature. For niobium, we can draw upon the large body of literature on the hydrogen-impurity problem. It was pointed out by Alefeld¹⁴ that diffusion studies of the heavy isotopes of hydrogen yield an activation energy that is comparable to the splitting between the ground and first excited vibrational states, as measured by neutron scatter-

ing. Estimating the isotope effect in a harmonic-oscillator approximation gives the result that on going from H^+ to μ^+ the zero point of such an Einstein oscillator is raised to a level close to the first excited state of H^+ . Therefore, the μ^+ in its lowest vibrational state is likely to achieve a tunneling rate between interstitial sites exceeding the 10^{12} -sec⁻¹ jump rate observed for H^+ at room temperature.

To explain the sharp line at the average field in our Nb data, a diffusion constant on the order of 10^{-2} cm²/sec is needed.¹⁵ With such a large diffusion constant at low temperature, moreover, it would appear that the muon might more properly be described as a propagating particle. Such behavior was speculated to exist for H^+ , although quantum mobility of H^+ has not been reported.¹⁶⁻¹⁸ For a bandlike model, a mean velocity on the order of 10^2 cm/sec is also consistent with our niobium results.

Trapping mechanisms could also affect diffusion. A possible explanation of the lead-indium results could include separate contributions from mobile muons with a diffusion constant of 10^{-4} cm²/sec and others in randomly located traps. Considering that materials-dependent parameters, such as the tunneling rate and mean free path, and the effects of defect trapping are expected to be important, further speculation lies beyond the scope of present information on our specimens. Further work to clarify the physical processes involved in this new species of ionic tunneling¹⁹ is planned.

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¹M. L. G. Foy, N. Heiman, W. J. Kossler, and C. E. Stronach, Phys. Rev. Lett. **30**, 1064 (1973). Data-acquisition techniques used in this reference were also used in the present experiments.

²J. H. Brewer, K. M. Crowe, F. N. Gyax, and A. Schenck, in "Muon Physics," edited by V. W. Hughes and C. S. Wu (Academic, New York, to be published).

³P. Pincus, A. C. Gossard, V. Jaccarino, and J. H. Wernick, Phys. Lett. **13**, 21 (1964); J. M. Delrieu and J. M. Winter, Solid State Commun. **4**, 545 (1966); A. G. Redfield, Phys. Rev. **162**, 367 (1967).

⁴H. Ullmaier, Comments Solid State Phys. **5**, 81

(1973); J. D. Cribier, B. Jacrot, L. Madhav Rao, and B. Farnoux, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland, Amsterdam, 1967), p. 161.

⁵J. Alonso and L. Grodzins, in *Hyperfine Structure and Nuclear Radiations*, edited by E. Matthias and D. A. Shirley (North-Holland, Amsterdam, 1968), p. 549.

⁶T. R. Brown and J. G. King, Phys. Rev. Lett. 26, 969 (1971).

⁷U. Essmann and H. Trüble, Phys. Lett. 27A, 156 (1968); U. Essmann, Phys. Lett. 41A, 477 (1972).

⁸I. G. Ivantaer and V. P. Smilga, Zh. Eksp. Teor. Fiz. 54, 559 (1968) [Sov. Phys. JETP 28, 286 (1968)]. This reference was pointed out to us by J. Brewer after our experiments had begun. Professor Brewer has discovered some errors in the calculations therein.

⁹J. Schelton, H. Ullmaier, and G. Lippmann, Z. Phys. 253, 219 (1972); H. W. Weber, J. Schelton, and G. Lippmann Phys. Status Solidi (b) 57, 515 (1973).

¹⁰J. Schelton, H. Ullmaier, and W. Schmatz, Phys. Status Solidi (b) 48, 619 (1973).

¹¹The thicknesses, residual resistance ratios, and Ginzburg-Landau parameters for the $\text{Pb}_{0.90}\text{In}_{0.10}$ and Nb specimen, respectively, are 2.7 and 3.2 mm; 5 and 20; 2.7 and 1.2.

¹²H. W. Weber and R. Riegler, Solid State Commun. 12, 121 (1973).

¹³I. I. Gurevich, E. A. Mel'eshko, I. A. Muratova, B. A. Nikol'sky, V. S. Roganov, V. I. Selivanov, and B. V. Sokolov, Phys. Lett. 40A, 143 (1972).

¹⁴G. Alefeld, in *Vacancies and Interstitials in Metals, Proceedings of an International Conference, Jülich, Germany, 1968*, edited by A. Seeger *et al.* (North-Holland, Amsterdam, 1969), p. 959. See also Ber. Bunsenges. Phys. Chem. 76, No. 8 (1972).

¹⁵The diffusion constant is estimated to be on the order of $\gamma_{\mu}^2 \langle \Delta B^2 \rangle A / \Delta \omega$, where $\langle \Delta B^2 \rangle$ is the mean square width of the internal field distribution, A the area of the vortex lattice cell, and $\Delta \omega$ the observed linewidth.

¹⁶A. Widom, Phys. Rev. B 4, 1697 (1971).

¹⁷G. Blaesser and J. Peretti, in *Proceedings of the International Conference on Vacancies and Interstitials in Metals, Jülich, Germany, 1968* (Kernforschungsanlage Jülich GmbH, Jülich, Germany), Vol. 2, p. 886.

¹⁸C. P. Flynn and A. M. Stoneham, Phys. Rev. B 1, 3966 (1970); A. M. Stoneham, Ber. Bunsenges. Phys. Chem. 76, 816 (1972).

¹⁹Tunneling states of defects in solids is reviewed by V. Narayanamurti and R. O. Pohl, Rev. Mod. Phys. 42, 201 (1970).