Anisotropy of the Magnetic-Field–Induced Phase Transition in Superconducting UPt_3

A. Schenstrom, M-F. Xu, Y. Hong, D. Bein, M. Levy, and Bimal K. Sarma
Department of Physics, University of Wisconsin–Milwaukee, Milwaukee, Wisconsin 53201

S. Adenwalla, Z. Zhao, T. Tokuyasu, D. W. Hess, J. B. Ketterson, and J. A. Sauls
Department of Physics and Astronomy, Northwestern University, Evanston, Illinois 60208

D. G. Hinks
Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439
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Measurements of the attenuation of longitudinal ultrasound in superconducting UPt_3 show a peak, below H_{c2}, which depends strongly on the orientation of the field relative to the c axis. We propose an explanation of this peak in terms of a vortex phase transition in an unconventional superconducting state, which is consistent with recent neutron-scattering data.

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Ultrasonic attenuation measurements have been performed in UPt_3 with both longitudinal and shear waves.[1-4] Both the observed power-law temperature dependence and the anisotropy in the attenuation of shear waves suggest an unconventional superconducting state in UPt_3. A curious λ-shaped peak in the temperature dependence just below T_c (Ref. 2) has also been observed.

Recently, Qian *et al.*[4] and Müller *et al.*[5] observed a peak in the magnetic field dependence of the ultrasonic attenuation. Measurements with shear waves propagating in the basal plane[6] failed to show this peak. These observations prompted immediate speculation that the peak is associated with a phase transition[4]; either a structural transition in which case the symmetry of the flux lattice changes, or a vortex-core transition in which the symmetry and structure of the order parameter changes within the core of each vortex, analogous to the vortex-core transition in superfluid ^{3}He-B.[7] Either scenario is *a priori* plausible if the superconducting state of UPt_3 is described by an unconventional order parameter. Evidence of a phase transition in a magnetic field has also been seen with a torsional oscillator.[8]

In this Letter we report extensive measurements that demonstrate the following:

(1) The attenuation peak in the magnetic field is observed with longitudinal sound propagating in the basal plane as well as along the c axis (no peak is observed for shear waves).

(2) There is a large anisotropy in the field strength H_{FL} of the above-mentioned peak. H_{FL} decreases as the field is tilted away from the c axis.

(3) The λ peak just below T_c is also seen with longitudinal sound propagating in the basal plane as well as along the c axis (once again, no peak is observed for shear waves).

(4) There are two additional features in the attenuation data in the superconducting state. A broad shoulder versus magnetic field, above H_{FL}, and another shoulder versus temperature, below the λ peak.

We have made measurements on two different samples, referred to as 1 and 2. The starting materials were somewhat better for sample 2. The transition temperatures T_c (measured inductively) were 508 and 530 mK, with transition widths of 30 and 15 mK, respectively; the second sample was clearly better. The attenuation change between the normal and superconducting state was larger in sample 2. In the first sample we observed only the field peak at H_{FL}. The second sample showed the λ peak and the two shoulders, versus temperature and magnetic field, respectively, in addition to the field peak.

In Fig. 1, we show the attenuation of ultrasound in temperature sweeps at zero field for sample 2. The data for $\mathbf{q} \parallel \mathbf{c}$ clearly show a peak in the attenuation just below T_c, and a shoulder at lower temperatures. Neither of

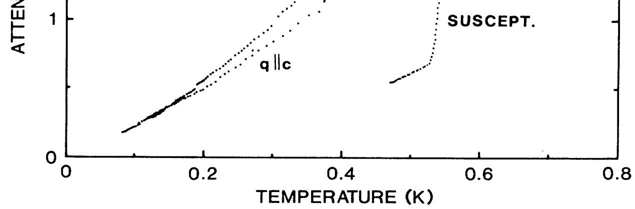

FIG. 1. Temperature dependence of the attenuation of longitudinal sound.

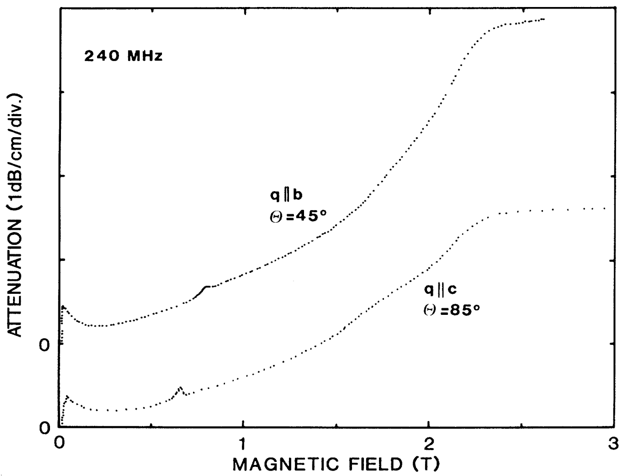

FIG. 2. Magnetic field dependence of the attenuation for two different propagation directions and angles of the magnetic field with respect to the c axis. The peak in the attenuation occurs at $H_{FL}=0.65$ T (0.78 T) for $\mathbf{q}\parallel\mathbf{c}$ and $\theta=85°$ ($\mathbf{q}\parallel\mathbf{b}$, $\theta=45°$).

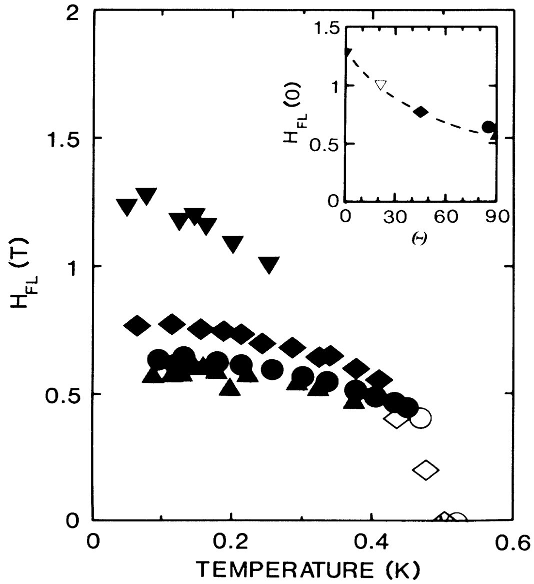

FIG. 3. H-T plot of $H_{FL}(T)$ (filled symbols) and the λ peak (open symbols), for different orientations of the magnetic field with respect to the c axis. (▼: 0°; ◆,◇: 45°; ●,○: 85°; ▲: 90°). Inset: the anisotropy of $H_{FL}(0)$.

these features are as clear for $\mathbf{q}\parallel\mathbf{b}$ at this frequency (240 MHz), but the λ peak was present at low frequencies (25 and 75 MHz), and the shoulder was observed at a higher frequency (460 MHz). The temperature dependence of the attenuation below 0.4 K (below the shoulder) can be fitted by $\alpha \sim T^n$, with $n \approx 1.2$ (1.3) for $\mathbf{q}\parallel\mathbf{c}$ ($\mathbf{q}\parallel\mathbf{b}$).

In Fig. 2, we plot the attenuation (again for sample 2) versus magnetic field (θ measured from the c axis, in the a-c plane). The temperature was 70 mK for $\mathbf{q}\parallel\mathbf{b}$ and 140 mK for $\mathbf{q}\parallel\mathbf{c}$. The difference in field at the peak ($H_{FL}=0.65$ and 0.78 T) is due to anisotropy, and not the difference in temperature. We identify the large increase in attenuation at very low fields (< 20 mT) with heating due to flux motion close to H_{c1} ($\sim$10 mT).[9]

Above H_{c2} ($\sim$2.0 T along the c axis) the attenuation flattens out. However, below H_{c2} there is again a broad shoulder, which is more clearly evident with sound propagating along the c axis. The change in attenuation $\alpha(H_{c2})-\alpha(0)$ is larger for $\mathbf{q}\parallel\mathbf{b}$ than for $\mathbf{q}\parallel\mathbf{c}$. A smaller attenuation difference for the two propagation directions was seen in the temperature data (Fig. 1). The larger difference versus magnetic field is due to the field dependence of the normal-state attenuation. This was verified in fields up to 8 T.

In Fig. 3 each symbol represents a complete run: the filled symbols are measured values of H_{FL} obtained from field sweeps at constant temperature. The open symbols are the positions of the λ peak obtained in a temperature sweep at constant field. Measurements on sample 1, with $\mathbf{q}\parallel\mathbf{H}\parallel\mathbf{c}$, are shown as ▼ [the extrapolated value $H_{FL}(T=0)$ was the same for sample 2 in this orientation]. The other three sets of data were all measured on sample 2: ◆ and ● are for $\mathbf{q}\parallel\mathbf{b}$, H at 45°, and $\mathbf{q}\parallel\mathbf{c}$, H at 85°, angles measured from the c axis in the a-c plane; ▲ is for $\mathbf{q}\parallel\mathbf{H}\parallel\mathbf{b}$. The inset of Fig. 3 shows the anisotropy of the peak in attenuation; $H_{FL}(T=0)$ is shown as a function of angle $\theta=\cos^{-1}(\hat{\mathbf{H}}\cdot\hat{\mathbf{c}})$, and is a factor of 2 larger along the c axis than in the basal plane. The measurement of Ref. 5 (▽) fits nicely with our data.

Although the two curves, H_{FL} from field sweeps and T_λ from temperature sweeps, may merge as suggested in Ref. 5, we are hesitant in identifying these two phenomena as the same. At a field >0.2 T, the λ peak is substantially broadened and the same is true for the field peak at temperatures above $\sim$350 mK. The errors in both H_{FL} and T_λ, close to the point where the two curves would meet, are therefore too large to make this assertion.

In addition, there are the two broad features (shoulders), seen below T_λ in temperature sweeps and above H_{FL} in field sweeps, respectively. Although we could not track these features versus temperature and field (because they are so broad), they may be indications of additional phase boundaries in the superconducting state. If we postulate that there are no more than two phase boundaries, we have to connect the shoulder seen in the field sweeps with the λ peak, and the shoulder seen in temperature sweeps with the field peak, for the phase boundaries not to cross. It should be noted that the two phase boundaries would have to come very close to each other at $\sim$0.5 T and 450 mK, which is also very close to $H_{c2}(T)$.

Although there is considerable literature on the possibility of unconventional pairing in UPt_3,[10] the symmetry class of the order parameter is not known with certainty. Using neutron-scattering measurements of the dynamic spin susceptibility, several authors[11] have constructed semiphenomenological models in which the pairing interaction is mediated by the exchange of antiferromagnetic spin fluctuations. These authors agree that such a pairing mechanism leads to an unconventional even-parity (singlet) state in UPt_3. In particular, Putikka and Joynt[12] predict that the order parameter belongs to the two-dimensional representation E_{1g} of D_6, and may be written in the form

$$\Delta(\mathbf{k}) = \Delta_0[\eta_1\theta(\mathbf{k}) + \eta_2\xi(\mathbf{k})] ,$$

where (η_1,η_2) are complex amplitudes that transform as a vector, $\boldsymbol{\eta} = \eta_1\hat{\mathbf{x}} + \eta_2\hat{\mathbf{y}}$, in the plane perpendicular to the c axis, and $\{\theta(\mathbf{k}),\xi(\mathbf{k})\}$ are the basis functions of E_{1g} given in Table I of Ref. 12. We discuss the Ginzburg-Landau (GL) theory for vortex states in UPt_3 based on this order parameter.

Starting from the free-energy functional of Serene and Rainer,[13] it is straightforward to derive the GL functional,

$$\Delta\Omega(\boldsymbol{\eta},\mathbf{A}) = \int d^3x[\alpha|\boldsymbol{\eta}|^2 + \beta_1|\boldsymbol{\eta}|^4 + \beta_2|\boldsymbol{\eta}\cdot\boldsymbol{\eta}|^2] + \int d^3x[\kappa_1(\nabla_i\eta_j)(\nabla_i\eta_j)^* + \kappa_2(\nabla_i\eta_i)(\nabla_j\eta_j)^* + \kappa_3(\nabla_i\eta_j)(\nabla_j\eta_i)^*]$$
$$+ \int d^3x\left[\kappa_4(\nabla_z\eta_i)(\nabla_z\eta_i)^* + \frac{|\boldsymbol{\partial}\times\mathbf{A}|^2}{8\pi} - \frac{\mathbf{H}\cdot(\boldsymbol{\partial}\times\mathbf{A})}{4\pi}\right] ,$$

where $\mathbf{A}$ is the vector potential, $\mathbf{H}$ is the external field, and $\boldsymbol{\nabla} \equiv \boldsymbol{\partial} - i2e\mathbf{A}/\hbar c$ is the covariant derivative. The coefficients in this functional are determined by Fermi-surface averages of the basis functions $\{\theta,\xi\}$ and the Fermi velocity $\mathbf{v}(\mathbf{k})$. Below T_c, $\alpha = N(E_F)\ln(T/T_c) < 0$, the order parameter in zero field is determined by minimizing the spatially homogeneous functional. Since stability requires $\beta_1 > 0$, the state that minimizes the GL functional depends upon the sign of β_2. For $\beta_2 < 0$, the order parameter is $\boldsymbol{\eta} \sim \hat{\mathbf{x}}$ or $\boldsymbol{\eta} \sim \hat{\mathbf{y}}$ (Ref. 14) with amplitude $|\boldsymbol{\eta}|^2 = |\alpha|/2(\beta_1+\beta_2)$, whereas $\beta_2 > 0$ leads to the complex order parameter $\boldsymbol{\eta}_+ = (|\alpha|/2\beta_1)^{1/2}(\hat{\mathbf{x}} + i\hat{\mathbf{y}})$, or the degenerate state $\boldsymbol{\eta}_- = \boldsymbol{\eta}_+^*$, as predicted in Ref. 12. These solutions are important since the weak-coupling (BCS) theory predicts that both $\beta_1,\beta_2 > 0$.[15]

Using the GL theory for the two-dimensional representations of D_6, Volovik[16] noted that the order parameter at the upper critical field H_{c2}, for fields along the c axis, is proportional to $\boldsymbol{\eta}_+ \sim \hat{\mathbf{x}} + i\hat{\mathbf{y}}$; there is no degeneracy at H_{c2} with the complex-conjugate solution. He further proposed that the peak in the acoustic attenuation be identified with a transition from the state $\boldsymbol{\eta} \sim \hat{\mathbf{x}}$ (or $\boldsymbol{\eta} \sim \hat{\mathbf{y}}$), which he assumed to be stable in zero field, to the state $\boldsymbol{\eta}_+ \sim \hat{\mathbf{x}} + i\hat{\mathbf{y}}$, close to H_{c2}. At high fields $H > H_{\mathrm{FL}}$, a triangular flux lattice is expected. In contrast, Volovik's assumed zero-field solution, $\boldsymbol{\eta} \sim \hat{\mathbf{x}}$, breaks axial symmetry about the c axis, implying that the vortex lattice in low fields, $H_{c1} < H < H_{\mathrm{FL}}$, is a distorted triangular lattice of singly quantized flux lines. Volovik further argued that the transition should disappear in a tilted field. Thus, the observation of the acoustic attenuation peak for fields perpendicular to the c axis seems to rule out his proposed transition and suggests that, if E_{1g} is the correct symmetry class, the zero-field solution is one of the complex phases $\boldsymbol{\eta}_\pm$, consistent with the prediction of Ref. 12.

We propose an alternative explanation for the field-dependent transition that is consistent with the low-field solution of Ref. 12. We have solved the GL equations for a single vortex in the background phase $\boldsymbol{\eta}_+$ (and $\boldsymbol{\eta}_-$). For the field oriented along the c axis, we find that, over a sizable region of the GL phase diagram, the minimum-energy vortex solution exhibits a spontaneously broken axial symmetry in which the vortex core has

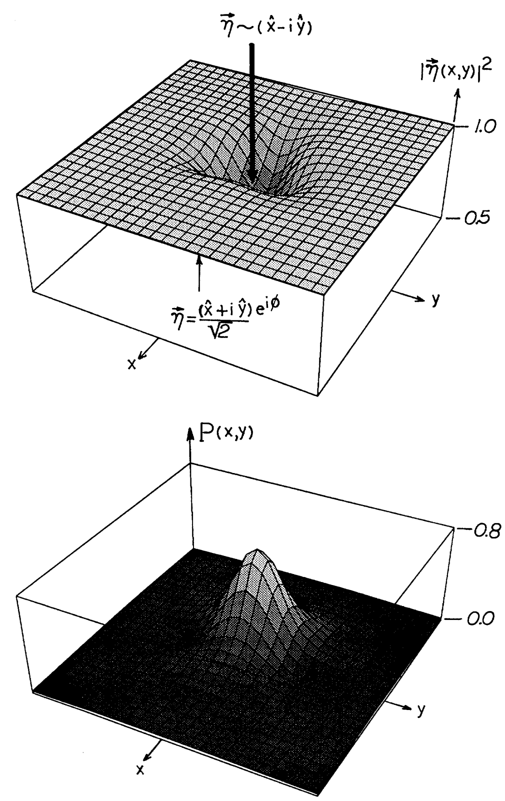

FIG. 4. The structure of a single vortex in the background phase $\boldsymbol{\eta}_+$. The upper figure shows the density $n_s = |\boldsymbol{\eta}(x,y)|^2$, which is finite everywhere. Inside the core, $\boldsymbol{\eta}$ is primarily given by the complex-conjugate phase $\boldsymbol{\eta}_-$. The projection of the $\boldsymbol{\eta}_-$ phase is shown in the lower figure: $P(\mathbf{x}) = |\boldsymbol{\eta}_- \cdot \boldsymbol{\eta}(\mathbf{x})|$. The grid size is one coherence length, ξ_{GL}.

lower symmetry than C_6. The asymmetry of the vortex core is clearly indicated by the density $n_s \sim |\boldsymbol{\eta}(\mathbf{x})|^2$, shown in Fig. 4. The triangular vortex core has dimensions of roughly $6\xi_{\rm GL}$, where $\xi_{\rm GL} = (\kappa_1/|\alpha|)^{1/2}$ is a GL coherence length, and has a nonvanishing superfluid density. Most importantly, the order parameter inside the core is not the phase $\boldsymbol{\eta}_+$, but rather the complex-conjugate phase $\boldsymbol{\eta}_-$. We find a stable solution to the GL equations with this structure for $0 < \beta_2/\beta_1 \leq 0.30$. For larger values of β_2/β_1 we obtain only axially symmetric solutions with a normal vortex core, i.e., $n_s = 0$ at the center of the vortex.

The existence of vortices in the order parameter $\boldsymbol{\eta}_+$ with a core of the complex-conjugate phase $\boldsymbol{\eta}_-$ is important for the evolution of the mixed state in high magnetic field. As $H \rightarrow H_{c2}$, and vortices become closely packed, the dominant superconducting phase is that of the core, i.e., $\boldsymbol{\eta}_-$, unless there is a vortex-core transition. This must be the case, since precisely at H_{c2} the lowest-energy solution of the linearized GL equations (determined by the gradient terms) is proportional to $\boldsymbol{\eta}_+$, which has different symmetry than that of $\boldsymbol{\eta}_-$. It is clear that the transition is induced by the vortex-vortex interactions[17] and occurs when the vortices become closely spaced. Qualitatively, we estimate the transition field to occur when the intervortex spacing is comparable to the size of the core, i.e., when $d \simeq 6\xi_{\rm GL}$ or $H_{\rm FL} \sim H_{c2}/\sqrt{6}$. There is no *a priori* reason for this transition to abruptly disappear when the field is tipped away from the c axis, although the vortex structure and order parameter are expected to show considerable anisotropy. However, more work is required to establish whether or not this vortex-core transition is correctly identified with the peak observed in high-field ultrasound studies.

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[1]D. J. Bishop, C. M. Varma, B. Batlogg, E. Bucher, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. **53**, 1009 (1984).

[2]V. Muller, D. Maurer, E. W. Scheidt, C. Roth, K. Luders, E. Bucher, and H. E. Bommel, Solid State Commun. **57**, 319 (1986).

[3]B. S. Shivaram, Y. H. Jeong, T. F. Rosenbaum, and D. G. Hinks, Phys. Rev. Lett. **56**, 1078 (1986).

[4]Y. J. Qian, M-F. Xu, A. Schenstrom, H-P. Baum, J. B. Ketterson, D. Hinks, M. Levy, and B. K. Sarma, Solid State Commun. **63**, 599 (1987).

[5]V. Müller, Ch. Roth, D. Maurer, E. W. Scheidt, K. Luders, E. Bucher, and H. E. Bommel, Phys. Rev. Lett. **58**, 1224 (1987).

[6]B. S. Shivaram, Y. H. Jeong, T. F. Rosenbaum, D. G. Hinks, and S. Schmitt-Rink, Phys. Rev. B **35**, 5372 (1987).

[7]P. J. Hakonen, M. Krusius, M. M. Salomaa, J. T. Simola, Yu M. Bun'kov, V. P. Mineev, and G. E. Volovik, Phys. Rev. Lett. **51**, 1362 (1983).

[8]R. N. Kleiman, Bull. Am. Phys. Soc. **33**, 215 (1988).

[9]A. Schenstrom, M-F. Xu, Y. Hong, M. Levy, B. K. Sarma, S. Adenwalla, Z. Zhao, J. B. Ketterson, and D. Hinks, in Proceedings of the Eighteenth Rare Earth Conference, Lake Geneva, Wisconsin, 12-16 September 1988 (to be published).

[10]D. Rainer, Phys. Scr. **T23**, 106-112 (1988).

[11]J. E. Hirsch, Phys. Rev. Lett. **54**, 1317 (1985); K. Miyake, S. Schmitt-Rink, and C. M. Varma, Phys. Rev. B **34**, 6554 (1986); M. R. Norman, Phys. Rev. Lett. **59**, 232 (1987).

[12]W. Putikka and R. Joynt, Phys. Rev. B **37**, 2372 (1988), and to be published.

[13]J. W. Serene and D. Rainer, Phys. Rep. **101**, 221 (1983).

[14]The fourth-order functional is minimized by any real vector $\boldsymbol{\eta}$. This orientational degeneracy is resolved in sixth order.

[15]In fact, weak-coupling theory implies the much stronger result, $\beta_2/\beta_1 = \frac{1}{2}$, independent of the specific choice of E_{1g} basis functions.

[16]G. E. Volovik, J. Phys. C **21**, L221 (1988).

[17]This transition is qualitatively different from the vortex-core transition in superfluid ^{3}He-B where the vortices are always so far apart that the vortex core is not influenced by interactions with neighboring vortices.