Mechanical Measurements of the Flux Lattice in the Heavy-Fermion Superconductor UPt_3

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We report mechanical measurements of the flux lattice in UPt_3. In the superconducting state the flux lattice that is formed in the sample when a magnetic field is applied contributes a restoring force and a dissipation term to a high-Q mechanical oscillator, when the oscillator and sample are tilted with respect to the static magnetic field. In addition to the characteristic response for a type-II superconductor that gives $H_{c2}(T)$, we observe an unexpected dissipation peak along two lines in the (H,T) plane within the superconducting state. The evidence suggests transitions between three different superconducting phases, and hence a complex phase diagram for UPt_3 is proposed.

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Soon after the discovery of the heavy-fermion superconductors, it was recognized that they represent a new class of materials in which the pairing mechanism for superconductivity may not be of the conventional phonon-mediated, singlet BCS type.[1] Considerable subsequent work suggest that these materials are likely to be superconductors with anisotropic higher-angular-momentum pairing states.[2] For example, in UPt_3 the pairing is thought to be *d* wave.[3] With higher-angular-momentum pairing, the possibility exists of transitions between different superconducting states, as in superfluid ^{3}He. Furthermore, the anisotropic nature of the superconducting state in these materials suggests that the flux lattice formed when a magnetic field between H_{c1} and H_{c2} is applied will have a complex character. The symmetry of the flux lattice can depend in a complex way on the density of the flux lines (i.e., the applied magnetic field) and the underlying symmetry of the order parameter. The cores of the flux lines themselves can, in general, be anisotropic depending on the orientation of the magnetic field with respect to the order parameter, which we presume to be pinned to the crystalline axes in the hexagonal UPt_3. For example, in ^{3}He, which is the only known anisotropic superfluid state, transitions between phases with singly and doubly quantized vortices, symmetric and asymmetric, core structures, and singular and nonsingular core structures have been considered theoretically[4] and investigated experimentally.[5] The observation of analogous transitions between different superconducting states or textures would be certain evidence of higher-angular-momentum pairing in the heavy-fermion superconductors.

In this paper we present results on the flux lattice in UPt_3 using a novel high-Q mechanical oscillator technique.[6,7] We observe dissipation peaks indicative of a complex phase diagram with three distinct superconducting phases in the (H,T) plane. Our work complements that of Müller *et al.*[8] and Qian *et al.*[9] who observe an attenuation peak in high-frequency ultrasound measurements of UPt_3 along the higher-field phase line reported in this paper. In addition, we observe a feature above $H_{c2}(T)$ which we believe is due to flux penetration into the superconducting sheath, at the higher field of $H_{c3}(T)$. We find that $H_{c3} \approx 1.7 H_{c2}$ in agreement with the standard theory of Saint-James and de Gennes[10] developed for isotropic superconductors.

In our apparatus a large single crystal of UPt_3 ($\sim$0.5 cm^3) is affixed to a high-Q BeCu mechanical oscillator, as shown schematically in the inset to Fig. 3. The $\hat{\mathbf{c}}$ axis of the crystal, the applied magnetic field $\mathbf{H}$, and the rotational axis of the oscillator are all parallel to within $\sim 2°$. ac susceptibility coils placed around the sample allow us to independently measure T_c. The resonant frequency ($\sim$600 Hz) and dissipation of the oscillator are measured in the experiment. In a type-II superconductor, the applied magnetic field produces a flux lattice in the sample for $T < T_c$ and $H_{c1} < H < H_{c2}$. The force and dissipation associated with tilting the crystal with respect to the static magnetic field are reflected as small changes in the oscillator response. The oscillator has a Q of $\sim 10^6$ with a frequency resolution of 1 part in 10^8. This gives high sensitivity to the small changes in mechanical damping and restoring force due to the presence of the flux lattice. Changes in the damping and restoring force of the oscillator signify the transitions at H_{c1}, H_{c2}, and H_{c3} and between the three superconducting phases. It is the superconducting phase diagram of UPt_3 which is the subject of this paper. A more detailed discussion of the mechanical oscillator technique for the study of type-II superconductors can be found in Ref. 7.

The additional restoring force on the crystal in the presence of the applied magnetic field is due to the pinning of the flux lines to the crystal.[11] In the limit of strong pinning, the pinned flux lines give rise to an effective dynamic sample magnetization because the rate of flux penetration through the surface barrier is slow as compared to the oscillator frequency. When operated in the torsional mode, this results in a restoring torque $\tau = MH\sin^2\theta_0 = BH\sin^2\theta_0$ on the oscillator, where θ_0 is the angle between the rotational axis of the oscillator $\boldsymbol{\omega}$

and the applied magnetic field **H**. If **H** and **ω** were perfectly aligned ($\theta_0=0$), there would be no restoring torque, whereas $\theta_0=90°$ gives too large a response for convenient measurements with our apparatus. In our experimental configuration, $\theta_0 \sim 2°$ is chosen in order to keep the damping and changes in frequency low, allowing a sensitive high-Q measurement over the whole range of H and T. This choice is also crucial in order to keep the measurement in the low-amplitude limit, i.e., where the amplitude of the flux-line motion (here ~10 Å) is small as compared to the lattice constant of the flux lattice. Large amplitudes of flux-line motion are found to mechanically melt the flux lattice that we wish to observe.

The additional dissipation term is due to the relative motion of the unpinned flux lines with respect to the crystal. There is a viscosity associated with the relative motion due to microcurrents which are driven through the normal cores, as calculated by Stephen and Bardeen.[12] We have verified that the oscillator response is linear with drive amplitude; thus we can be sure that the damping is viscous and not in a hysteretic regime at the amplitude of operation. In the strong-pinning limit, which holds well at low fields and temperatures, $(\Delta f/f) \propto BH$ and there is no dissipation because all of the flux lines are pinned to the crystal. Near H_{c2} the flux pinning begins to weaken and finally vanishes at H_{c2}. Consequently, the restoring force from the pinned flux lines vanishes at H_{c2}. As the flux lines depin, the free vortices give rise to dissipation since they move relative to the crystal lattice. Just below H_{c2} there is a dissipation peak when the majority of flux lines depin. Above H_{c2} the dissipation is due to the eddy-current damping of the normal metal.

Shown in Fig. 1 are the real and imaginary parts of the oscillator response as a function of applied magnetic field at fixed temperature (upper) and as a function of temperature at fixed magnetic field (lower). Looking first at the lower figure, we see that in the superconducting state there is an additional stiffness (~50 ppm) associated with the flux pinned inside the sample. The damping (Q^{-1}) is low and is due to the residual eddy-current damping of the normal-metal oscillator. As the flux lattice melts at $T_c(H)$, the real part of the response softens, accompanied by a large dissipation peak in the imaginary component. In the normal region ($T > T_c \sim 0.53$ K) eddy-current damping is large, but the real part of the response vanishes since the magnetic field penetrates uniformly throughout the sample. The dissipation *peak* occurs at H_{c2} as measured by the ac susceptibility. We believe that the shoulder above the peak is due to H_{c3} and will be discussed below. In the upper figure the response as a function of magnetic field at fixed temperature is shown. The real part of the response is quadratic in field as discussed above. However, if one divides out the quadratic field dependence (as done in the upper figure) then the field dependence of the normalized response is qualitatively similar to the temperature dependence of the response shown in the lower figure.

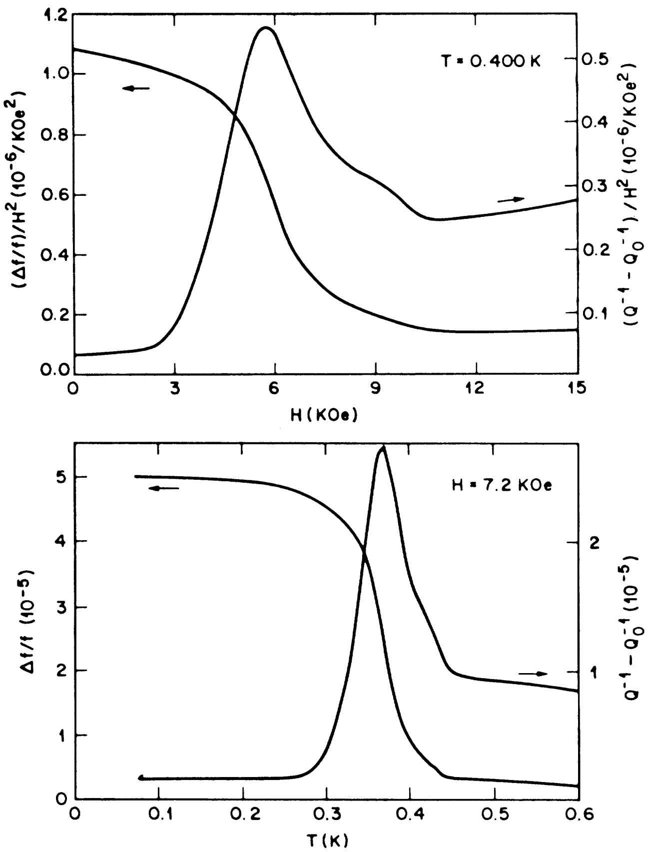

FIG. 1. Top: The normalized frequency shift and dissipation of the oscillator with the UPt_3 sample attached, at fixed temperature ($T=0.400$ K) as a function of magnetic field with $\mathbf{H} \parallel \hat{c}$. We identify the large peak as H_{c2} and the shoulder as H_{c3}. Bottom: The frequency shift and dissipation at fixed magnetic field ($H=7.2$ kOe) as a function of temperature. The features are the same as in the upper figure.

Shown in Fig. 2 is the normalized dissipation on a more sensitive scale as a function of magnetic field at fields below H_{c2}. At the lowest temperature (0.076 K) we observe two dissipation peaks *below* H_{c2} which we believe indicate transitions between three distinct superconducting phases. Measurements on conventional superconductors show no such additional structure in the superconducting state.[7] The dissipation peaks are small compared to the one observed at H_{c2}, and cannot be seen easily at the scale shown in Fig. 1. No corresponding feature is observed in the real part of the response. As the temperature is increased the peaks move together, and at 0.3 K and 5.5 kOe they meet at a tricritical point. We have checked carefully for hysteresis and all the

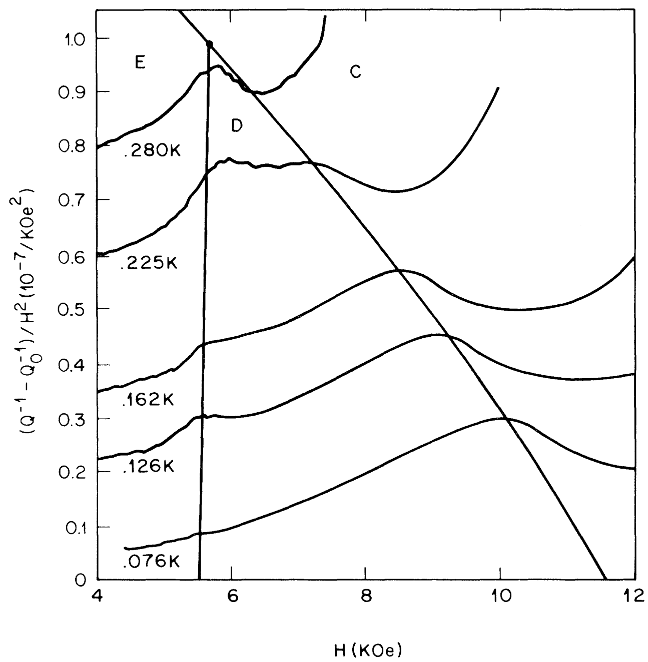

FIG. 2. The normalized dissipation of the oscillator at a series of temperatures as a function of field below H_{c2}, on an expanded scale. The offsets between curves corresponding to different temperatures are arbitrary. The attenuation peaks indicate transitions between three distinct superconducting phases, which meet at a tricritical point (0.3 K and 5.5 kOe). The lines are merely a guide to the eye. It is the peak positions that are plotted in Fig. 3.

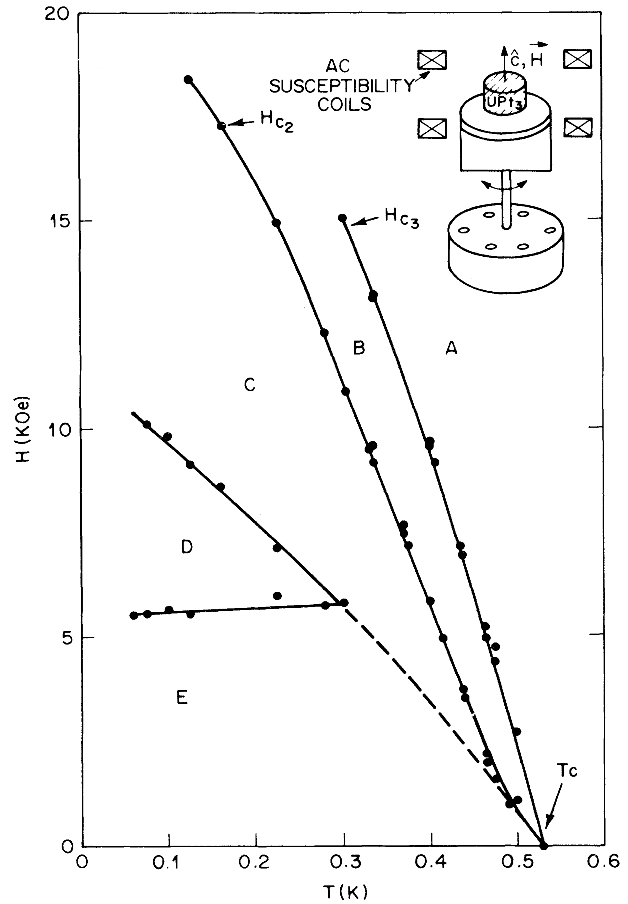

FIG. 3. Proposed phase diagrams for UPt_3. A: normal metal; B: sheath superconductivity; C, D, and E: superconducting states. The dashed line connecting the tricritical point and T_c is suggested to complete the phase diagram. A schematic drawing of the experimental apparatus is shown in the upper right-hand corner.

transitions appear to be second order or only weakly first order. Shown in Fig. 3 is our proposal for the superconducting phase diagram of UPt_3. The dashed line in the phase diagram is based on the assumption that the upper phase line is proportional to $H_{c2}(T)$ and hence meets the H_{c2} line at T_c. It is interesting to note that the change in the character of the transition from the solid line to the dashed line is also observed in the ultrasound measurements.[8]

In our opinion, the three bulk superconducting phases are most likely to be ones with different symmetries of the flux lattice. In an isotropic superconductor with large κ, the flux lattice is triangular[13] at all fields below $H_{c2}(T)$ and temperatures[9] below T_c. However, the difference in free energy between the square and triangular lattices is quite small and the calculation must be done to second order to obtain the proper equilibrium configuration. Since the energy balance is so delicate it is quite likely that in an anisotropic superconducting state the symmetry of the flux lattice will reflect the underlying symmetry of the order parameter. For example, cubic anisotropy is known to stabilize a square flux lattice in low-κ conventional superconductors.[14] Our model is that the observed dissipation peaks correspond to transitions between superconducting phases with flux lattices of different symmetry. The additional dissipation in the neighborhood of the transition arises because of the motion of the flux lines relative to each other and to the crystal lattice, as the flux lattice changes symmetry. The fact that there is no softening of the real part of the response at these transitions, as at $H_{c2}(T)$, confirms the fact that the dissipation peaks observed cannot simply be attributed to monotonic changes in the pinning forces.

Recently, Müller *et al.*[8] and Qian *et al.*,[9] in ultrasound measurements, have also seen evidence for a complex phase diagram in UPt_3. They observe an attenuation peak along one phase line which is similar to that which we find between region C and the combined areas of D and E. The fact that there is no significant frequency dependence to the position of the phase line over six decades of frequency is strong evidence that the dissipation peaks observed in *both* measurements are due to a true phase transition in the superconducting state and not a simple artifact of flux pinning. However, they find no evidence for a tricritical point. Müller *et al.*[8] find evidence in the susceptibility for a hysteretic phase at low fields within our region E. Their measurements were made with **H** tilted with respect to the $\hat{\mathbf{c}}$ direction by 21°. We find no evidence for such a phase in our experiment with $\mathbf{H} \parallel \hat{\mathbf{c}}$. Clearly, it would be important to investigate the phase diagram of UPt_3 with the magnetic field oriented perpendicular to the $\hat{\mathbf{c}}$ direction, in order to compare to theories for different anisotropic superconducting states.

In Fig. 1 there is a shoulder in the real and imaginary parts of the oscillator response above $H_{c2}(T)$ which we believe to be the response due to H_{c3}. In a type-II superconductor, the critical field for the destruction of sheath superconductivity is higher than H_{c2} because of specular reflection of the quasiparticles at the interface.[10] This makes it harder to insert a quantum of flux into a region the size of a coherence length and superconductivity is not destroyed until one reaches a field larger than H_{c2} which is denoted as H_{c3}. The position of the observed feature is plotted in Fig. 3 and tentatively denoted as H_{c3}.

Saint-James and de Gennes[10] have calculated H_{c3} for an isotropic superconducting state and find $H_{c3} = 1.695 H_{c2}$ in good agreement with experiments in conventional superconductors.[15] Our measurements agree well with this value giving $H_{c3} \approx 1.7 H_{c2}$. In general, one would expect this relationship to be modified for an anisotropic state. However, recent measurements suggest that the most likely superconducting gap structure for UPt_3 is one with a line of nodes in the basal plane.[3] This would produce isotropic vortices for the case of the magnetic field parallel to the $\hat{\mathbf{c}}$ axis (our case). Seen in that light the agreement with isotropic theory is not surprising. Clearly, measurements with different field orientations will prove interesting as the theory of Saint-James and de Gennes will require modification for anisotropic superconducting states.

In conclusion, we have studied the properties of the flux lattice in UPt_3 using a novel mechanical oscillator technique. We find evidence for a complex phase diagram with three distinct superconducting phases. The phase diagram we propose differs from that derived from the ultrasound measurements in the observation of an additional phase line at intermediate fields. We suggest that the phases which we see represent states with different symmetries of the flux lattice. However, it is also possible that the observed transitions are occurring instead in the cores of the individual flux lines. Clearly, neutron-scattering experiments are called for to elucidate the nature of these phases and their symmetry.

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[1]C. M. Varma, in *Moment Formation in Solids,* edited by W. J. L. Buyers (Plenum, New York, 1984), and Bull. Am. Phys. Soc. **29**, 404 (1984); P. W. Anderson, Phys. Rev. B **30**, 1549 (1984).

[2]D. J. Bishop, C. M. Varma, B. Batlogg, E. Bücher, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. **53**, 1009 (1984); Z. Fisk, H. R. Ott, T. M. Rice, and J. L. Smith, Nature (London) **320**, 124 (1986), and references therein.

[3]K. Miyake, S. Schmitt-Rink, and C. M. Varma, Phys. Rev. B **34**, 6554 (1986); S. Schmitt-Rink, K. Miyake, and C. M. Varma, Phys. Rev. Lett. **57**, 2575 (1986).

[4]A. L. Fetter, in *Progress in Low Temperature Physics,* edited by D. F. Brewer (North-Holland, Amsterdam, 1986), Vol. 10, p. 1.

[5]P. L. Gammel, T.-L. Ho, and J. D. Reppy, Phys. Rev. Lett. **55**, 1708 (1985).

[6]See, for example, R. N. Kleiman, G. K. Kaminsky, J. D. Reppy, R. Pindak, and D. J. Bishop, Rev. Sci. Instrum. **56**, 2088 (1985), and references therein.

[7]R. N. Kleiman, P. L. Gammel, E. Bücher, and D. J. Bishop, to be published.

[8]V. Müller, Ch. Roth, D. Maurer, E. W. Scheidt, K. Lüders, E. Bücher, and H. E. Bömmel, Phys. Rev. Lett. **58**, 1224 (1987).

[9]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).

[10]D. Saint-James and P. G. de Gennes, Phys. Lett. **7**, 306 (1964).

[11]B. H. Heise, Rev. Mod. Phys. **36**, 64 (1964).

[12]M. J. Stephen and J. Bardeen, Phys. Rev. Lett. **14**, 112 (1965); John Bardeen and M. J. Stephen, Phys. Rev. **140**, A1197 (1965).

[13]D. Criber, B. Jacrot, L. Madhau Rao, and B. Farnoux, in *Progress in Low Temperature Physics,* edited by C. J. Gorter (North-Holland, Amsterdam, 1967), Vol. 5, p. 161.

[14]B. Obst, Phys. Lett. **28A**, 662 (1969).

[15]See, for example, B. Serin, in *Superconductivity,* edited by D. Parks (Dekker, New York, 1969), Vol. 2, p. 969.