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Inductive response of oriented UPt_3 in the superconducting state

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The inductive response of single crystals of UPt_3 have been studied from 1 K to 70 mK using mutual inductance techniques to measure χ' and χ'' at 31.7 and 317 Hz and tunnel diode resonant methods at 3 and 16 MHz, for the excitation fields parallel to the symmetry axis of the crystal. A double bump in the χ'' measurements is clearly distinguishable and is consistent with the $H=0$ phase diagrams of other workers. The temperature dependence of the magnetic penetration depth has been extracted from the measurements which indicate that the low-frequency results are linear for $T/T_c < 0.5$, while the high-frequency data indicate a power-law dependence close to T^2.

Following the discovery of superconductivity in the heavy-fermion system of UPt_3,[1] attention has been focused on identifying the nature of the superconducting state.[2] Although earlier measurements indicated that the superconducting gap did not cover the entire Fermi surface, the case for unconventional superconductivity strengthened with the observation of a double jump at T_c in the specific heat,[3] with the identification of the multiple regioned phase diagram,[4–12] and with the interpretation of H_{c2} studies.[13–15] The magnetic-field penetration depth, $\lambda(T)$, of the superconducting state at low temperatures may be helpful in the identification of the ground state since $\lambda(0)$ provides the ratio n_s/m, where n_s is the condensate density and m is the mass. A completely gapped Fermi surface would lead to the usual behavior where the change in $\lambda(T)$ is $\Delta\lambda(T) \sim \exp[-\Delta_0/T]$. A superconductor possessing nodes in the gap structure would have $\Delta\lambda(T) \sim T^n$, where n is a number of order unity that depends on the crystallographic direction being studied and the symmetry of the superconducting state. Due to this sensitivity to the crystallographic orientation, $\lambda(T)$ provides information not available from the specific heat.

Several measurements of $\lambda(T)$ in UPt_3 have been reported,[16–22] and each is inconsistent with an isotropic gap for the superconducting state. Conventional magnetization techniques, whether dc or ac, have the advantage that the relative uncertainties can be extremely small. However, the experiments are often sensitive to the structure of the surface and are not capable of obtaining $\lambda(0)$ with a single measurement. In principle, the muon spin relaxation (μ^+SR) technique is ideally suited for the problem. The field distribution that μ^+SR probes is not sensitive to the crystal surface, and $\lambda(T)$ itself, rather than just $\Delta\lambda(T)$, emerges directly from the analysis. The main disadvantage, when compared to the magnetization measurements, is the relatively low precision. Despite this difficulty, the results of Broholm *et al.*[20] were the first to reveal an anisotropy in $\lambda(T)$ and led the authors to conclude that superconducting UPt_3 exists in a hybrid state.[23] However, the interpretations of the μ^+SR results for UPt_3 are controversial.[21] The experiments are usually performed in fields much larger than H_{c1}, where the field distribution in the sample should be independent of the applied field, thus allowing for extraction of the London penetration depth. Unfortunately, measurements in the high-field regime do not reveal any change upon entering the superconducting state, either because $\lambda(0)$ is too long [the field distribution $\langle \Delta B^2 \rangle \sim (1/\lambda^4)$], or because of the presence of a strong temperature-dependent background.[21]

The purpose of this work was to measure $\lambda(T)$ using inductive techniques, with the same samples over a broad frequency range (31 Hz–16 MHz). Our goal was to check whether the previous results, obtained by several groups using different samples, could be understood in a single framework. We have now investigated a variety of UPt_3 samples provided by different sources. While a detailed description of all of our work will be given in another paper,[24] the present paper will report the results of our study of the highest-quality specimens. These single crystals were grown at Los Alamos using a bismuth flux from which the crystals were quenched. Two needles, with approximate dimensions 6 mm long and a diameter of 1–2 mm were used in our inductive studies. After annealing at 950 °C in vacuum for 100 hours, the specimens were etched in a 3:1 solution of HCl:HNO_3 and washed. Using a standard optical microscope, the surface of the needles appeared uniform and defect-free to at least the 50-μm scale.

In order to characterize our samples, a standard four-wire ac resistivity measurement was made on one of the the two needles that were studied inductively, and the results are shown in Fig. 1. The needlelike morphology limited us to measurements along the c axis, for which we observed the normal-state resistivity to follow a temperature dependence typical for heavy-fermion materials, namely $\rho(T) = \rho(0) + AT^2$, with $\rho(0) = 0.6 \pm 0.1$ $\mu\Omega$ cm, $A = 0.7 \pm 0.1$ $\mu\Omega$ cm K^{-2}, and $\rho(300\ \text{K})/\rho(0) = 260$. The uncertainty in $\rho(0)$ follows from geometric considerations. These values, along with the relatively high-

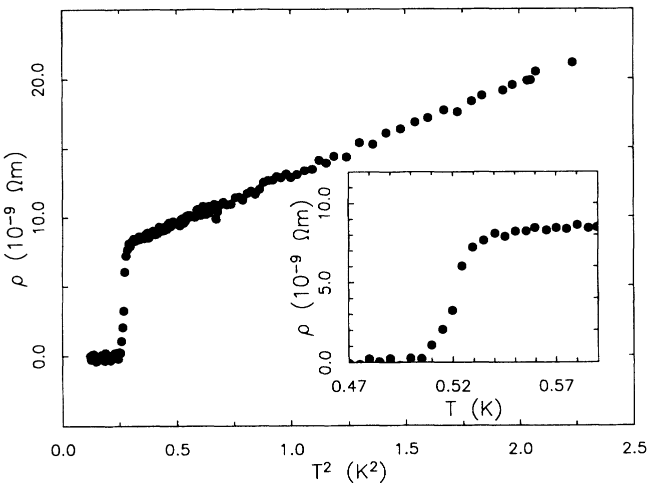

FIG. 1. The resistivity ρ is shown as a function of T^2. The inset shows ρ vs T in the vicinity of T_c, where T_c(midpoint) = 520 mK. A fit of the normal-state data to $\rho(T)=\rho(0)+A\,T^2$ yields $\rho(0)=0.6\pm0.1$ $\mu\Omega$ cm and $A=0.7\pm0.1$ $\mu\Omega$ cm K^{-2}.

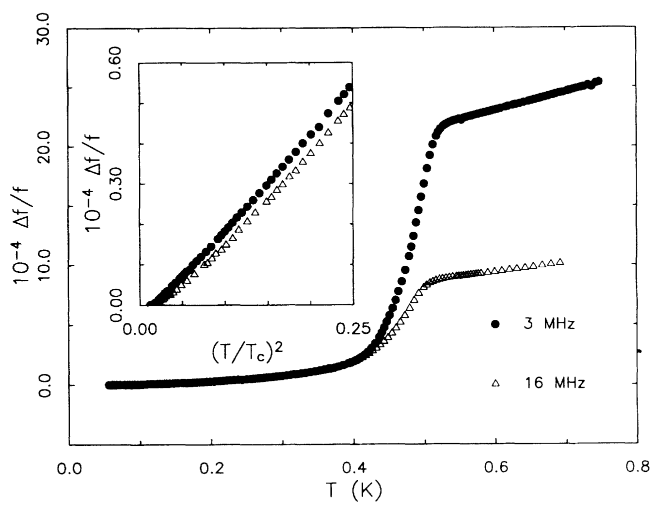

FIG. 2. The relative rf frequency response $\Delta f/f \equiv \{f(0)-f(T)\}/f(T)$ is shown as a function of T for two nominal frequencies of 3 and 16 MHz. The inset shows the $(T/T_c)^2$ dependence for $T/T_c<0.5$.

superconducting transition at 520 ± 10 mK, indicate that the quality of the single crystals is very good.

The high-frequency (3 and 16 MHz) inductive measurements were made using a tunnel diode resonator technique.[25,26] The samples are placed in an inductor ($\hat{\mathbf{H}}\parallel\hat{\mathbf{c}}$) that is 8 mm long with a bore size of 3 mm, where the inductor is part of an inductance-capacitance LC tank circuit. The relative change in resonant frequency of the circuit is given by

$$\frac{\Delta f}{f}=-\frac{1}{2}\left[\frac{\Delta L}{L}\right]\left\{1-\frac{2}{Q^2}\right\}-\frac{1}{Q^2}\left[\frac{\Delta R}{R}\right], \qquad (1)$$

where R is the effective resistive component and Q is the quality factor. At the low temperatures of this study, changes in inductance result from the response of the sample only. In the normal state, the response is determined by the classical skin depth, $\delta=\{2/(\mu\sigma\omega)\}^{1/2}$, where μ is the permeability and σ is the low-frequency conductivity. Far below T_c, changes in the resonant conditions of the circuit are generated by $\Delta\lambda(T)$ which, for an s-wave superconductor, would be frequency independent for $\omega\ll\Delta_0$. The results of our tunnel diode measurements on the UPt_3 samples are shown in Fig. 2. For both frequencies, the resonant conditions have also been studied with the samples removed, and in each case, the background temperature dependence is negligible.

In the normal state, assuming an infinitely long coil-sample geometry, the leading order term in Eq. (1) gives

$$\frac{\Delta f}{f}=-\frac{1}{2}\left[\frac{\Delta L}{L}\right]=-\frac{1}{2}\left[\frac{2\pi R}{A}\right]\Delta\lambda, \qquad (2)$$

where R is the radius of flux exclusion of the sample and A is the total area containing the flux. In the superconducting state, Eq. (1) may be written as

$$\frac{\Delta f}{f}=-\frac{1}{2}\left[\frac{\Delta L}{L}\right]=-\frac{1}{2}\left[\frac{2\pi R}{A}\right]\Delta\lambda, \qquad (3)$$

which is nearly exact since $(\Delta R/R)\to0$ and $Q>100$ for our circuit. Our measurement of $\Delta f/f$, for two frequencies, and $\rho(T)$ allows us to use Eq. (2) to determine $\lambda(T\to0)$ since the field penetration at T_c is $\delta(\omega,T_c)$. This analysis gives $\lambda(T\to0)=1.3$ μm when using the ρ that we measured along the c axis. However, Shivaram, Rosenbaum, and Hinks[13] and de Visser *et al.*[27] have systematically studied the resistivity along the c axis and in the a-b plane for a variety of samples from different sources. These workers found both $\rho(0)$ and A, with $\rho(T)=\rho(0)+AT^2$, in the a-b plane to be approximately 2 to 3 times larger than when measured along the c axis. Therefore, when multiplying our measured $\rho(0)$ and A by a factor of 3, our analysis yields $\lambda(T\to0)=2.1$ μm. This value, which we consider to be an upper bound, is larger than that reported by some workers[19,20] but is consistent with others,[17] and within the limit of $\lambda(T\to0)>1.1$ μm placed by Uemura *et al.*[21]

Our low-frequency (31.7 and 317 Hz) ac susceptibility measurements were made using standard mutual inductance techniques with $\hat{\mathbf{H}}\parallel\hat{\mathbf{c}}$. Two PAR 124A lock-in amplifiers were used to simultaneously measure both χ' and χ'' signals. The results at 317 Hz are shown in Fig. 3, and although the signal to noise was not as good, the 31.7 Hz data gave essentially the same results. The ac field strength is $\lesssim100$ mG, and results similar to those shown in Fig. 3 were also obtained with a threefold increase in ac field strength. The signal of the empty coil has been studied, and the temperature dependence of the background signal is negligible. A striking feature of Fig. 3 is the clear identification of a double bump associated with the two superconducting transitions, which is consistent with the $H=0$ phase diagram of other workers.[3-9] The absolute phase is difficult to set, but it may also be adjusted numerically after data acquisition.[24] A small adjustment of 1.5° is enough to zero the normal-state χ'' contribution without affecting the presence of the double bumps or the low T/T_c dependence of χ' to be discussed below. A small nonzero contribution would be expected from the normal-

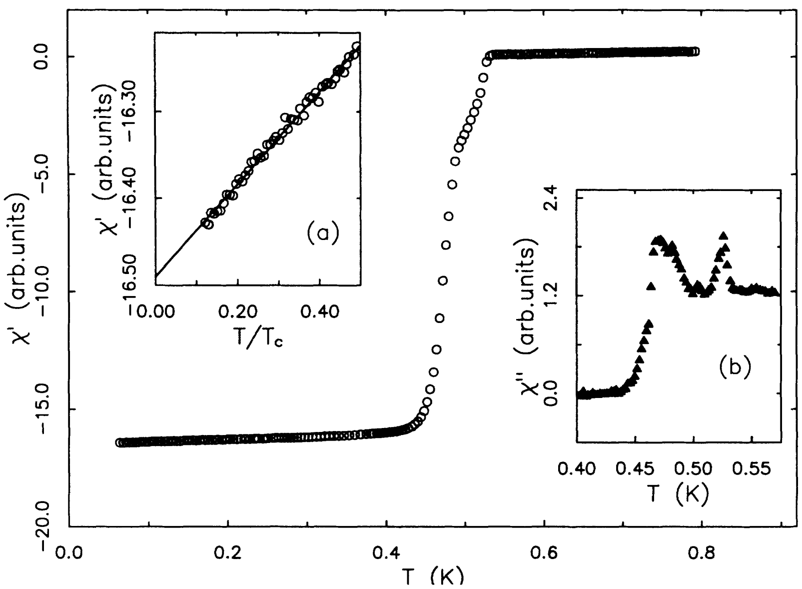

FIG. 3. The real, χ', low-frequency (317 Hz) response in arbitrary units is shown as a function of T. The inset (a) shows the (T/T_c) dependence of χ' for $T/T_c < 0.5$. The inset (b) shows the imaginary, χ'', response in the vicinity of T_c.

state conductivity.

The insets in Figs. 2 and 3(a) show the temperature dependence of a quantity which is directly proportional to $\Delta\lambda(T)$. For the rf work, Fig. 2, the temperature dependence is approximately T^2 for $T/T_c < 0.5$, while the low-frequency work is linear in T/T_c in the same regime. An explanation for this difference might include the response from flux penetration into the bulk of the sample. However, the low-frequency results are independent of ac excitation amplitude and reproduce with temperature cycling provided the sample does not warm above $T \sim 0.6\ T_c$. While the tunnel diode circuit does not have a continuously variable amplitude, the estimated field amplitude ($H \sim 100$ mG) is extremely small, and the same reproducibility as for the low-frequency measurements is observed. Therefore, we consider this explanation unlikely. We also exclude the possibility that the rf measurements are more sensitive to the real part of the complex impedance of the superconductor (i.e., surface resistance R_s), which has been studied by Grimes, Adams, and Bucher.[28]

Clearly, our observations are inconsistent with an activated temperature dependence, i.e., $\Delta\lambda(T) \sim \exp[-\Delta_0/T]$. In addition, our results are different from those reported by several other workers. First, the penetration depth from dc magnetization studies for $\hat{\mathbf{H}} \parallel \hat{\mathbf{c}}$ has been reported to be T^2 (Refs. 16–18); however, this temperature dependence is expected when significant impurity scattering is present.[17,29,30] Second, other rf measurements have indicated a T^4 dependence which is independent of orientation;[19] however, not all the data in this work goes to very low temperatures and a significant amount of scatter in the data causes the temperature dependent identification to be more suggestive rather than conclusive.[24] We conclude that the data do indicate a frequency dependence of the penetration depth at low T/T_c, and note that our low frequency observation of a linear dependence of $\Delta\lambda(T)$ for $\hat{\mathbf{H}} \parallel \hat{\mathbf{c}}$ is consistent with the interpretations of some μ^+SR data.[20] The results are summarized in Fig. 4, with the 317-Hz data represented by the shaded region, where the upper and lower bounds have taken $\lambda(0)$ to be 1.3 and 2.1 μm, respectively. Geometric factors and the uncertainty of $\rho(T)$ in the basal plane prevent us from identifying the slope more precisely. However, these uncertainties do *not* compromise the linear temperature dependence shown in Fig. 3.

It has recently been pointed out by Putikka, Hirschfeld, and Wölfle[30] that a frequency-dependent penetration depth is possible for anisotropic superconductors. They

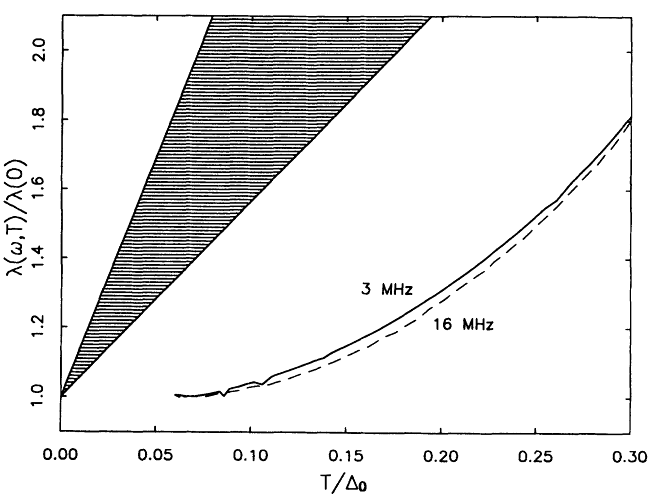

FIG. 4. The changes in the measured temperature and frequency-dependent penetration depth, $\lambda(\omega,T)$, normalized to its $T=0$ value, $\lambda(0)$, are plotted as a function of reduced temperature T/Δ_0, where Δ_0 has the BCS value of $1.76T_c$. The shaded region indicates a range of slopes possible for the low-frequency linear results, see text.

performed a model calculation for the E_{1g} state in the clean limit with the Poynting vector $\mathbf{q} \| \hat{\mathbf{c}}$ and with the screening currents $\mathbf{j}$ flowing in the basal plane, i.e., $\hat{\mathbf{H}} \perp \hat{\mathbf{c}}$. The calculations, however, do not apply directly to our experiments which would require both the $\mathbf{q}$ and $\mathbf{j}$ vectors to be in the basal plane. Furthermore, our samples are not in either the clean limit, $l \gg \xi_0$ (l is the mean free path), or the dirty limit, $l \ll \xi_0$, since $l \simeq \xi_0$.

In summary, we have inductively measured the magnetic field-penetration depth over the wide frequency range 31.7 Hz–16 MHz. The results indicate that there is a much stronger variation with temperature at low frequencies, $\Delta\lambda(T) \sim T$, than in the rf regime, where $\Delta\lambda(T) \sim T^2$. Interpreting the results in terms of either a screening enhancement from quasiparticle-quasihole pairs or impurity scattering is not presently possible.

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