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Logarithmic ac response in the heavy-fermion superconductor URu_2Si_2

P. Visani, Y. Dalichaouch, M. A. Lopez de la Torre, B. W. Lee, C. L. Seaman, and M. B. Maple
Department of Physics and Institute for Pure and Applied Physical Sciences, University of California, San Diego, La Jolla, California 92093
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ac-magnetic-susceptibility $\chi_{ac}(T)$ measurements as a function of frequency f and excitation field h_{ac} have been performed on a single crystal of the heavy-fermion superconductor URu_2Si_2 in magnetic fields H up to 30 kOe applied along the c axis. The apparent superconducting transition temperature T_c shows an unexpected and approximately logarithmic increase (decrease) with f (h_{ac}) that is enhanced with H, which suggests the existence of an irreversibility line below the upper critical field $H_{c2}(T)$ in URu_2Si_2. Isothermal χ_{ac} vs H sweeps were used to define $H_{c2}(T)$ which was independent of f and h_{ac}.

Heavy-fermion superconductors such as $CeCu_2Si_2$, UBe_{13}, UPt_3, and URu_2Si_2 have attracted considerable attention since they appear to exhibit anisotropic superconductivity, characterized by an energy gap that vanishes at points and/or lines on the Fermi surface, in which the electron pairing is mediated by a nonphonon mechanism.[1] Investigations of heavy-fermion superconductors in magnetic fields have been particularly informative. The upper critical field of UBe_{13} has been found to have an anomalous shape and the highest initial slope near T_c ever observed,[2] and features in $H_{c2}(T)$ of UPt_3 (Ref. 3) and URu_2Si_2 (Ref. 4) have been observed and attributed to the existence of at least two distinct superconducting transitions. The UPt_3 compound, which has a complex H-T phase diagram in which the two zero-field superconducting transitions converge to a critical point in higher field, has been the focus of a number of theoretical investigations in terms of d-wave superconductivity with singlet-spin pairing mediated by antiferromagnetic spin fluctuations.[5–8]

In this paper, we report ac susceptibility $\chi_{ac}(T)$ measurements as a function of frequency f and excitation field h_{ac} on a URu_2Si_2 single crystal in dc magnetic fields applied along the tetragonal c axis. We observe a logarithmic dependence upon f and h_{ac} of $\chi_{ac}(T)$ in URu_2Si_2 similar to what has been found in the high-T_c oxide superconductors and associated with the existence of an irreversibility line $H(T^*)$ below $H_{c2}(T)$ in the H-T plane. In addition, we have determined what appears to be the true upper critical field H_{c2} from field-swept isothermal χ_{ac} measurements.

The URu_2Si_2 specimen used in this study is a platelet-like single crystal, with approximate dimensions $2.0\times1.8\times0.2$ mm^3, cleaved from the center of a polycrystalline sample that was arc melted in an argon atmosphere and annealed in vacuum at 900 °C for 7 days. Back-scattered Laue x-ray diffraction analysis confirmed that the sample is a single crystal with the c axis perpendicular to the largest face. In-phase (χ') and out-of-phase (χ'') components of χ_{ac} were measured in a dilution refrigerator with a standard mutual inductance technique in the ranges $5\leq f\leq1000$ Hz and $0.004\leq h_{ac}\leq2.2$ Oe. The dc magnetic susceptibility χ_{dc} was measured down to 0.45 K using a Faraday magnetometer. A superconducting solenoid was used to apply dc magnetic fields up to 30 kG along the tetragonal c axis and parallel to h_{ac}.

Figure 1 shows the temperature T dependence of the real and imaginary parts χ' and χ'' of χ_{ac} measured with $H=0$. The data were taken for several values of h_{ac} between 0.004 and 2.2 Oe at $f=16$ Hz. Both components of χ_{ac} yield superconducting transition curves that shift to significantly lower temperatures with increasing values of h_{ac}. The possibility of edge effects in thin single crystals cannot be excluded, and the observation of fine structure in χ'' [Fig. 1(b)] measured in low fields ($h_{ac}<0.04$ Oe) might be an indication of such a contribution. Temperatures determined from the maximum slope of χ' and

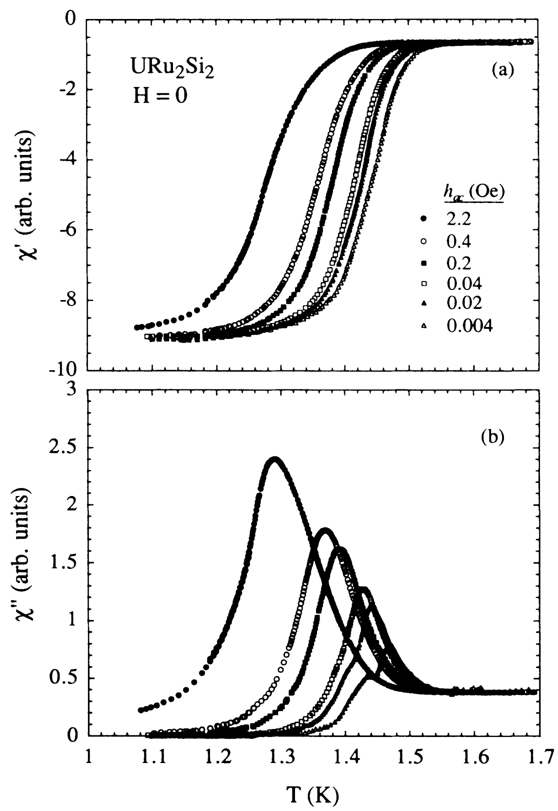

FIG. 1. Temperature dependence of the (a) inductive and (b) resistive components of the ac susceptibility measured in various ac fields at a frequency of 16 Hz for a URu_2Si_2 single crystal in zero applied dc magnetic field.

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from the maximum in χ'' (hereafter called T^*) decrease, first rapidly, and then less quickly at higher excitation fields. While the drop in χ' remains essentially unchanged, the peak in χ'' increases by more than 50% in magnitude and its width increases as h_{ac} is increased. These effects are not associated with Joule or eddy current heating since the primary coil of the probe is made of superconducting NbTi wire and no appreciable dependence of T_c on h_{ac} could be detected for an aluminum standard with $T_c = 1.16$ K, measured in the same run under identical conditions. The results for selected applied dc fields, shown in a semilogarithmic plot in Fig. 2(a), reveal that T^*, which is determined from the maximum in χ'', follows a roughly *logarithmic* decrease with increasing h_{ac}, an effect which is enhanced by magnetic field. The trend of the data over the applied field range shows a monotonic increase of the slope $dT^*/d\ln(h_{ac})$ with increasing H.

A striking feature is the frequency dependence of T^*, shown in Fig. 2(b) for $h_{ac} = 0.4$ Oe, where the logarithm of f is plotted vs $1/T^*$. The temperature T^*, determined from the maximum in χ'', increases logarithmically with increasing frequency, typically at a rate of 20 mK per decade of frequency, and the effect is enhanced by H as seen from the slopes of the lines. As a point of comparison, the T_c of the aluminum standard, measured in the same manner, did not reveal any appreciable frequency dependence. While these frequency dependences are small, $dT^*/d\ln(f) = 9$ mK in $H = 0$, they represent, nevertheless, 50% of the value estimated by Malozemoff *et al.* for Y-Ba-Cu-O crystals.[9] These unexpected results combined with the recent measurements of strong magnetic relaxation in $CeCu_2Si_2$ by Mota, Visani, and Pollini,[10] and UPt_3 by Pollini *et al.*,[11] strongly suggest that the H-T line extracted from these "transitions" does not represent the upper critical magnetic field $H_{c2}(T)$, which, of course, should be independent of f.

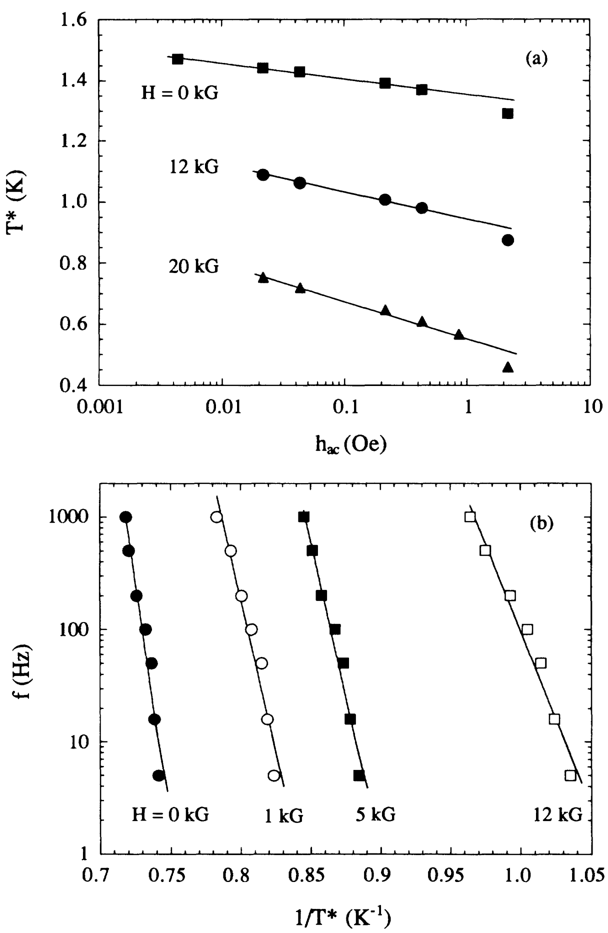

FIG. 2. (a) Dependence of the temperature T^* associated with the maximum in χ'' measured at frequency $f = 16$ Hz on excitation field h_{ac}, and (b) f vs $1/T^*$, illustrating the logarithmic dependence of T^* on f. The data were extracted from ac susceptibility measurements on a URu_2Si_2 single crystal for various applied fields $\mathbf{H} \| c$ and $h_{ac} = 0.4$ Oe.

The existence in high-T_c superconductors of a region below $H_{c2}(T)$ where flux motion is reversible has been the subject of an enormous amount of work over the past few years. Until recently, it was believed that the reversible region in the H-T plane for conventional superconductors is negligibly small and could not be detected experimentally because the onset of strong diamagnetism and superconducting critical currents occur pretty much along the same $H_{c2}(T)$ line. However, recent experiments by Suenaga *et al.*[12] have shown the existence of a surprisingly large reversible flux motion region in Nb_3Sn and Nb-Ti multifilamentary wires. Similar results were previously found in a single crystal of the Chevrel phase compound $PbMo_6S_8$ by Rossel *et al.*[13] who established the existence of an irreversibility line $H(T^*)$ below $H_{c2}(T)$ and observed strong magnetic relaxation. The existence of an appreciable reversible region in the H-T plane of low-temperature superconductors could be due to a combination of the material's superconducting parameters κ, $H_{c2}(T)$, and $\xi(T)$ which would broaden the thermal excitations of a liquidlike state to energies well below T_c and fields below H_{c2}.[14]

Flux lines in a type-II superconductor are subject to a driving force in the presence of an alternating modulation field $h_{ac}\cos\omega t$ which results in flux motion and subsequent energy losses. In an ideal pinning case where magnetic hysteretic losses dominate, the critical state model predicts a maximum in the temperature dependence of χ'' when the flux profiles due to the alternating magnetic field penetrate throughout the entire material. The penetration L_p of the flux inside the sample is constant and given by[15]

$$L_p = h_{ac}/J_c \tag{1}$$

where J_c is the critical current density. Within this framework, the temperature dependence of χ'' depends on the sample dimensions, J_c and h_{ac}, but is independent of f. For increasing h_{ac}, J_c must also increase according to Eq. (1) which implies that T^* must decrease as observed experimentally.

In the ideal flux-flow case, ac losses are dominated by the viscous motion of flux vortices and a maximum is also observed in χ'' when the ac field fully penetrates the sample. The flux-flow skin depth is determined by the flux-flow resistivity and the measuring frequency, but is independent of the ac field amplitude.[12] Dissipation due to viscous damping of vortex motion has been studied, recently, in the context of thermally activated flux flow by van der Beek and Kes[16] who found that the onset temperature T^* of irreversibility is related to f by

$$f \propto \exp(-U/k_B T^*) , \quad (2)$$

where U is the activation energy. This expression predicts a logarithmic dependence of the irreversibility temperature T^* on f as observed in Fig. 2(b). The similarities of these curves with the ones obtained from ac susceptibility and electrical resistivity measurements in high-T_c oxide superconductors by van der Beek and Kes[16] and Palstra et al.,[17] respectively, are, indeed, striking. A plot of $\ln(f)$ vs $1/T^*$ should therefore yield the same information on flux-motion pinning energy U as a plot of $\ln(\rho)$ vs $1/T$. Neglecting its intrinsic temperature dependence, U obtained from the slopes of Fig. 2(b) decreases for increasing fields with typical values of 18 meV at $H=0$ and 6 meV for $H=12$ kG. These values are comparable or even smaller than those found in high-T_c superconductors (i.e., $U=20-200$ meV in Y-Ba-Cu-O single crystals[18]). Our estimate of U in URu_2Si_2 is comparable to the value of 3 meV determined for a single crystal of $CeCu_2Si_2$, by Mota et al.,[19] from the decay of the remanent magnetization.

The simultaneous dependence of T^* on f and h_{ac}, shown in this work for URu_2Si_2, can be explained within the framework of a thermally activated flux-creep model which combines both pinning and flux flow of vortices. Using an extension of the flux-creep theory, Worthington et al.[20] derived an expression for the irreversibility line of the form

$$(1-T^*/T_c)^{3/2} = A \ln[(h_{ac}/f)F(H,T)] , \quad (3)$$

where A is a constant. If we neglect the temperature and field dependences of the argument of the logarithm, the above expression predicts an overall logarithmic dependence of the irreversibility line $H(T^*)$ both on f and h_{ac} amplitudes. From Fig. 3, one can see that the scaling behavior of the irreversibility line, in the form predicted by Eq. (3), is in qualitative agreement with our data. However, the observed ac field dependence appears to deviate from logarithmic behavior above $h_{ac} \approx 1$ Oe. Frequency and field dependences of the irreversibility line extracted from χ_{ac} measurements have been reported for Y-Ba-Cu-O by Malozemoff et al.[9] and confirmed by many other groups for different high-T_c single crystals and thin films. A study by Emmen, Stollman, and De Jonge[21] of epitaxial Y-Ba-Cu-O thin films also reveals f and h_{ac} dependences very similar to those found in this work. Identification of the peak position in χ'' with the irreversibility temperature T^* has been particularly controversial.[22] The argument is that χ_{ac} is a measure of ac field penetration into the sample and cannot be used to determine the irreversibility line except in the limit of low f and low h_{ac}.[23] New measurements on single crystals of high-temperature superconductors have also shown that data taken with a small ac field superimposed upon a larger dc field give good agreement with the irreversibility line $H(T^*)$ determined from dc magnetization measurements.[24] In this case, χ'' should peak at T^* and the ac measurements are sampling the magnetic state of the sample.

H vs T/T_c curves obtained from $\chi_{ac}(T,H)$ data taken at fixed frequency $f=16$ Hz and $h_{ac}=0.4$ Oe, where T_c is the critical temperature in zero applied dc field, are plotted in Fig. 4 for H$\|c$. Below 5 kOe, all the curves extracted from $\chi'(T)$ and $\chi''(T)$ data taken at various applied dc fields reveal positive curvature in low fields remarkably similar to the irreversibility line $H(T^*)$ observed in high-T_c oxide superconductors. When plotted in the form of $\ln(H)$ vs $\ln(1-T^*/T_c)$, the data deduced from χ'' show two regions that conform to a power-law dependence of $H(T^*)$ with an exponent that changes at $T^*/T_c=0.94$ and $H=0.2$ kOe from 1.3 in low fields to 4.2 up to 3 kOe. To determine the true upper critical field H_{c2} which should be independent of f and h_{ac}, we have used a different experimental approach. In dc magnetization measurements, H_{c2} can often be determined from a

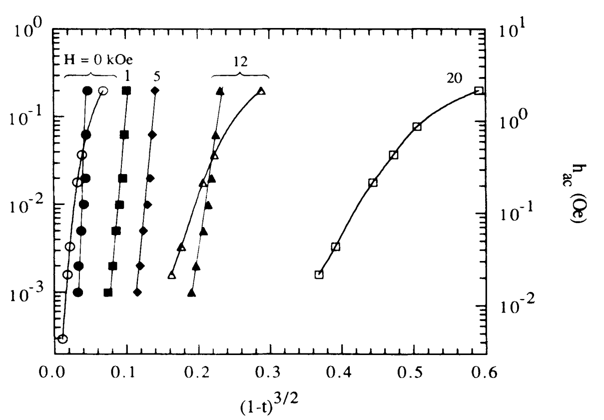

FIG. 3. $1/f$ (filled symbols) and h_{ac} (open symbols) vs $(1-t)^{3/2}$ for URu_2Si_2, where $t \equiv T^*/T_c$.

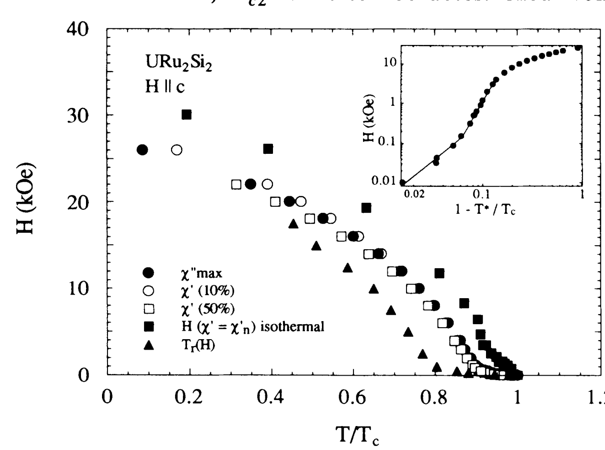

FIG. 4. H-T/T_c phase boundaries determined from $\chi'(T,H)$, $\chi''(T,H)$, dc magnetization, and isothermal $\chi'(H)$ data taken at $f=16$ Hz and $h_{ac}=0.4$ Oe on a URu_2Si_2 single crystal for H$\|c$. Open circles ($T_c=1.43$ K) and squares ($T_c=1.36$ K) represent the 10% drop and maximum slope in χ', respectively, whereas filled circles ($T_c=1.37$ K) indicate the maximum in χ''. Filled squares ($T_c=1.55$ K) are from isothermal χ' vs H sweeps, while filled triangles ($T_c=1.43$ K) are extracted from dc magnetization measurements. Inset: log-log plot of H vs $1-T^*/T_c$ data determined from $\chi''(T,H)$.

discontinuity in the slope of the reversible part of the magnetization M vs H as the hysteresis loop closes. However, in nonmagnetic or weakly magnetic superconductors, such as heavy-fermion compounds, this feature is hard to identify. In addition to the difficulty of determining H_{c2}, the problem inherent in the dc technique is one of sensitivity, especially for small samples. We have therefore made field-swept measurements of the in-phase component of χ_{ac} at constant temperature by stabilizing the field at each data point taken. The critical field H_{c2} is defined as the lowest dc field at which χ' coincides with the normal-state value, e.g., χ' at $T=1.6$ K. We observe that the value of H_{c2} measured in this way is independent of f and h_{ac} in the parameter window of this experiment as would be expected for a thermodynamic transition. The $H_{c2}(T)$ curve extracted from isothermal measurements should be equivalent to that extracted from isofield measurements provided T_c is defined as the onset of the transition in χ' or the absorption peak in χ''. We have chosen to use isothermal measurements of H_{c2} because the data showed less scatter than in the case of isofield measurements. In contrast to the line determined from the maximum in χ'' isofield measurements, the $H_{c2}(T)$ curve extracted from isothermal field-swept ac susceptibility measurements (Fig. 4) exhibits negative curvature down to 3 kOe, followed by a kink below which $H_{c2}(T)$ again shows slight negative curvature. The conventional method of determining the irreversibility line $T_r(H)$ is from dc magnetization measurements by defining T^* as the temperature where the zero-field cooled and field cooled magnetization curves separate. The results from such measurements on the URu_2Si_2 single crystal are shown in Fig. 4. The temperature dependence of $T_r(H)$ is in qualitative agreement with that of the H-T lines extracted from isofield χ' and χ'' measurements, in support of the interpretation that the real and imaginary parts of the ac susceptibility relate to the irreversibility line. However, the data extracted from dc magnetization lie below the data from $\chi'(10\%)$, $\chi'(50\%)$, and χ''_{max}.

These dependences of χ_{ac} on f and h_{ac} have important implications for experiments that attempt to determine $H_{c2}(T)$. While the region between the irreversibility line $H(T^*)$ and the true upper critical field curve $H_{c2}(T)$ might not occupy much phase space in the H-T plane, it can, nevertheless, yield misleading positive curvatures in $H_{c2}(T)$ near T_c, as shown in Fig. 4.

In conclusion, we have observed an unexpected and approximately logarithmic dependence of the critical temperature on the measuring frequency and excitation field in a single-crystal specimen of the heavy-fermion superconductor URu_2Si_2. This behavior is consistent with the existence of an irreversibility line $H(T^*)$ below the upper critical field $H_{c2}(T)$, similar to what has recently been observed in high-T_c oxide superconductors and some conventional superconductors. Isothermal field-swept ac susceptibility measurements, which are frequency and ac excitation field independent, apparently yield the true upper critical field $H_{c2}(T)$. However, even though our results are in qualitative agreement with predictions based on a thermally activated flux creep model, we cannot exclude the possibility that these unexpected ac susceptibility results are associated with fluxoid glass-lattice melting and/or unusual magnetic properties of URu_2Si_2.

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