Low-Temperature Susceptibility of the Noncentrosymmetric Superconductor $CePt_3Si$

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We report ac susceptibility measurements of polycrystalline $CePt_3Si$ down to 60 mK and in applied fields up to 9 T. In a zero applied field, a full Meissner state emerges at temperatures $T/T_c < 0.3$, where $T_c = 0.65$ K is the onset transition temperature. Though transport measurements show a relatively high upper critical field $B_{c2} \sim 4$–5 T, the low-temperature susceptibility χ' is quite fragile to the applied field, with χ' diminishing rapidly in fields of a few kG. Interestingly, the field dependence of χ' is well described by the power law $4\pi\chi' + 1 = (B/B_c)^{1/2}$, where B_c is the field at which the onset of resistance is observed in transport measurements.

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Two separate but related considerations are paramount in the search for superconductors with extraordinary electronic properties. The first is the role of crystal structure in determining the order parameter symmetry and the second is the nature of underlying electron correlations. As an example of the latter, the newly discovered itinerant ferromagnetic superconductors, represented by UGe_2 [1] and $ZrZn_2$ [2], are believed to exhibit a superconducting phase that is, in fact, mediated by a preemptive magnetic ordering. Antiferrromagnetic analogs are also known, such as UPd_2Al-3 [3] and the quasi-2D heavy fermion 1-1-5 family, $CeMIn_5$, where $M =$ Ir, Rh, or Co [4]. The superconducting phases in these two classes of systems are believed to be unconventional and are currently the subject of intense investigation [5]. More recently a new heavy fermion superconductor was reported in the itinerant antiferromagnet $CePt_3Si$ [6], which owing to its magnetic ground state, falls broadly into the same class as the 1-1-5 superconductors. The superconducting state in this material, however, is not only of interest in terms of magnetism, but also because it has a rather unusual crystal structure that lacks a center of inversion. A considerable amount of theoretical work has been done on two-dimensional (2D) systems lacking inversion symmetry, where the presence of a surface almost trivially guarantees the breaking of the symmetry [7]. In such systems spin-orbit scattering can have a significant impact on the spin states of the superconductor, forcing spins to lie in the plane, thereby destroying the spin isotropy. More generally, the effects of spin-orbit scattering are in some sense enhanced in noncentrosymmetric systems and will mix order parameter states of different parity. Similar arguments have been made for bulk systems with noninversion symmetry [8]. This suggests that $CePt_3Si$ is a compelling candidate for the realization of unconventional superconductivity mediated by the interplay between broken inversion symmetry, spin-orbit scattering, and magnetism [9]. Up to now only heat capacity and transport studies have been reported on $CePt_3Si$, though the antiferromagnetic order has been established via neutron scattering [10]. In the present Letter, we present low-temperature magnetic susceptibility measurements of polycrystalline $CePt_3Si$ samples and show that the material exhibits a full Meissner state well below the superconducting transition. Though believed to be a type-II superconductor, the zero temperature susceptibility obeys a power-law field dependence which is inconsistent with standard type-II behavior but is perhaps more characteristic of quantum criticality [11]. Furthermore, we show that upon doping La for Ce at the 2% level the superconducting transition is suppressed

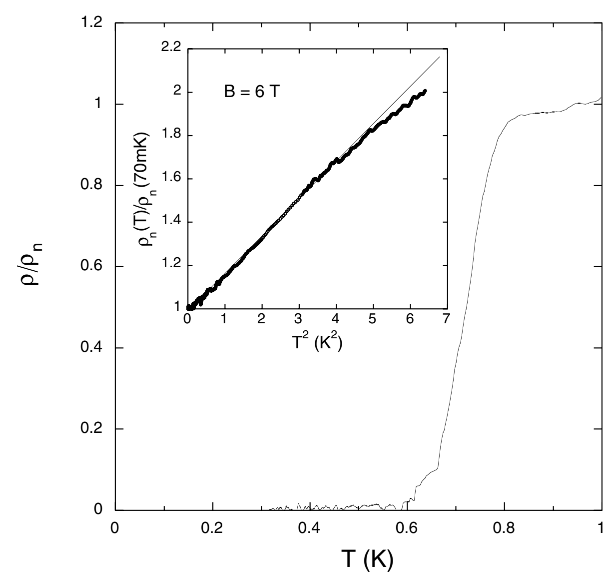

FIG. 1. Relative resistivity of $CePt_3Si$ as a function of temperature. The midpoint of the superconducting transition is at $T_c = 0.72$ K. Inset: quadratic temperature dependence of the normal state resistivity.

to below 50 mK. The implications of these observations for the nature of the pairing is discussed.

Polycrystalline samples were prepared by argon arc melting of stoichiometric quantities of high purity Ce, Pt, and Si. The resulting buttons were then annealed under vacuum at 900 °C for 5–10 days. The correct stoichiometry was verified via standard powder x-ray diffraction analysis. Scanning electron microprobe analysis of annealed samples did not show any evidence of impurity phases nor of Pt cluster formation. Transport measurements were made using a lock-in amplifier in a 4-wire configuration. Low-temperature ac susceptibility measurements were made with an astatically wound susceptibility coil operating at 505 Hz. Typical excitation fields were ~0.05 G, and external fields up to 9 T where applied parallel to the probe field. Transport and susceptibility measurements below 2 K were carried out on a dilution refrigerator with a base temperature of 50 mK. Measurements above 2 K were made using a Quantum Design Physical Properties Measurement System (PPMS) magnetometer.

In Fig. 1 we plot the normalized resistivity of $CePt_3Si$ as a function of temperature in zero magnetic field. The midpoint transition temperature $T_c = 0.72$ K and transition width $\Delta T_c/T_c \sim 0.15$ are in good agreement with the values reported in Ref. [6]. The residual resistance ratio $\rho(290\ \mathrm{K})/\rho(2\ \mathrm{K}) \approx 5$ was also comparable to that reported in Ref. [6]. In the inset of Fig. 1 we plot the low-temperature normal state resistivity as a function of T^2. These data were taken in an applied field of 6 T to suppress the superconducting state, and the linearity of the data is consistent with a Fermi liquid state. The solid line has a slope of 0.167 K^{-2}, which is about a factor of 3 smaller than that reported in Ref. [6]. However, the data in Ref. [6] were restricted to temperatures above T_c.

The real and imaginary components of the zero-field-cooled ac susceptibility, χ' and χ'', respectively, are plotted as a function of temperature in the main panel of Fig. 2. Note that a bulk superconducting state is observed, $4\pi\chi' \rightarrow -1$, with a corresponding dissipation peak in χ'' at a midpoint transition temperature $T_c = 0.56$ K. This is somewhat lower than that determined from the resistive transition in Fig. 1, but transport T_c's are often higher than their susceptibility counterparts, owing to the fact that susceptibility is a bulk probe, whereas resistivity is more sensitive to the emergence of continuous superconducting paths. The small bump in χ' between 0.4 and 0.5 K was evident in all of the samples studied, but its origin is unknown. In the inset of Fig. 2 we present χ' data above 2 K, where there is clearly a magnetic ordering transition at $T_N = 2.97$ K, which we believe is the Néel transition. The T_N in Fig. 2 is moderately higher than that obtained from the heat capacity data of Ref. [6].

We have examined the field dependence of the susceptibility by repeating the temperature scans shown in Fig. 2 in a variety of applied fields, as is shown in Fig. 3. Though we observed transient effects during the field sweeps, there was no significant hysteresis in the data. Zero-field cooling and field cooling produced the same values of χ' and χ''. The low-temperature diamagnetic response is extinguished on a field scale that is small in comparison to B_{c2} but large in comparison with B_{c1}. Though the transport critical field in $CePt_3Si$ is high, $B_{c2} \sim 4$–5 T, and, in fact, above the

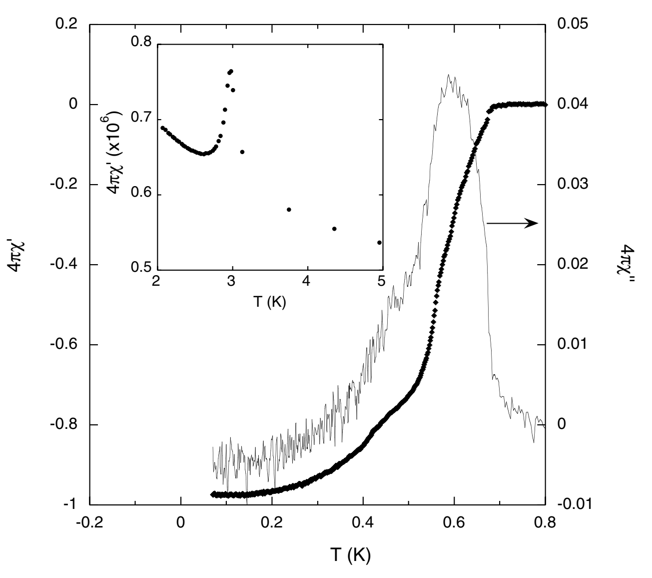

FIG. 2. Real and imaginary components of the ac susceptibility as a function of temperature in a zero applied field. The probe field was 0.05 G. Note that a full Meissner state is formed below ~0.3 K. Inset: ac susceptibility as a function of temperature showing an antiferromagnetic transition at $T_N = 2.97$ K.

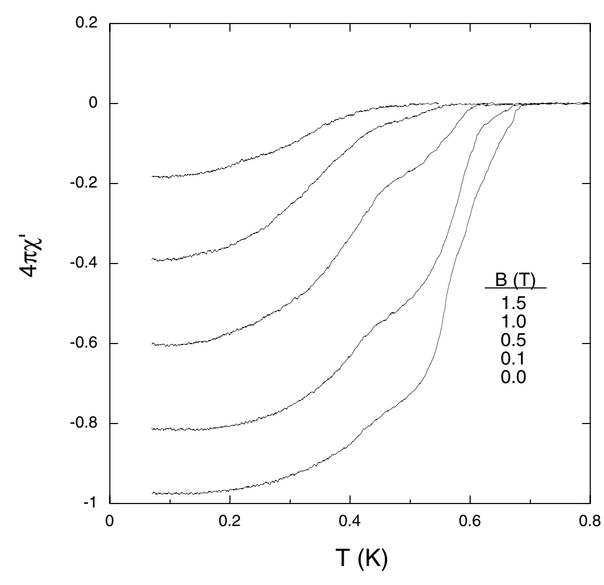

FIG. 3. Susceptibility as a function of temperature in an external magnetic field. Note the rapid increase in $4\pi\chi'$ upon the application of relatively modest fields. The field at which the diamagnetic response was completely attenuated was $B_c \sim 2.4$ T.

Clogston spin-paramagnetic limit [12], a field of only 0.5 T is sufficient to reduce the Meissner fraction by a factor of 2. On the other hand, the lower critical field of $CePt_3Si$ is believed to be quite small. The London penetration depth is estimated to be $\lambda_L \sim 1\ \mu$m and $\kappa \sim 140$ [6], which gives $B_{c1} \sim 1$ mT [13]. From this perspective it is surprising that a measurable Meissner fraction exists at a field $\sim 500 B_{c1}$ in a material with such a large value of κ.

The field dependence above B_{c1} exhibits power-law behavior as can be seen in the main panel of Fig. 4, where the symbols represent the susceptibility of two different $CePt_3Si$ samples. In this plot we have scaled the field axis by the field at which the diamagnetic response goes to zero, $B_c = 2.4$ T (see Fig. 5). The linearity of the data demonstrates unambiguously that the low-temperature susceptibility obeys a square-root scaling law, $4\pi\chi' = (B/B_c)^{1/2} - 1$, with a corresponding magnetization

$$4\pi M \sim -B[1 - 2/3(B/B_c)^{1/2}]. \tag{1}$$

In a classic type-II superconductor the vortex density above B_{c1} is $n \sim B/\Phi_o$, where Φ_o is the flux quantum. Because the vortex cores are normal, the Meissner fraction and corresponding diamagnetic magnetization usually decreases linearly with field up to B_{c2}, $4\pi M \sim (B - B_{c2})$. Interestingly, it has recently been shown that a square-root singularity, similar to that in Eq. (1), arises in the magnetization of ferromagnetic spin-triplet superconductors exhibiting a spontaneous flux lattice [14]. Though spontaneous vortex nucleation seems unlikely in a purely antiferromagnetic superconductor, if the applied field induces a ferromagnetic component to the ground state, then the mechanism described in Ref. [14] may play a role in the observed magnetization behavior. This would assume, of course, that $CePt_3Si$ is a spin-triplet superconductor. We also note that the mixed state of nodal superconductors is known to exhibit a $B^{1/2}$ anomaly in the subgap density of states [15], but the implications for the susceptibility are unclear. Nonetheless, the square-root behavior in Fig. 4 is highly unusual. For example, we have also plotted the 10-K susceptibility of a polycrystalline V_3Si sample as a function of $B^{1/2}$ in the inset of Fig. 4. V_3Si undergoes a second order structural phase transition just above its superconducting transition temperature $T_c = 17$ K, which breaks the inversion symmetry of the crystal [16]. Otherwise, it is believed to be a conventional superconductor. Note the V_3Si does not obey the square-root scaling, indicating that the scaling is not simply a consequence of the absence of inversion symmetry. It is evident from a comparison between the data in the inset and the main body of Fig. 4 that the susceptibility of $CePt_3Si$ is not representative of classic type-II behavior and almost certainly reflective of an unconventional superconducting ground state [17,18].

To the extent that itinerant antiferromagnetism mediates superconductivity in $CePt_3Si$, the critical field behavior may be driven by the field dependence of the underlying magnetic order parameter. Additional evidence for the importance of the antiferromagnetic ground state in

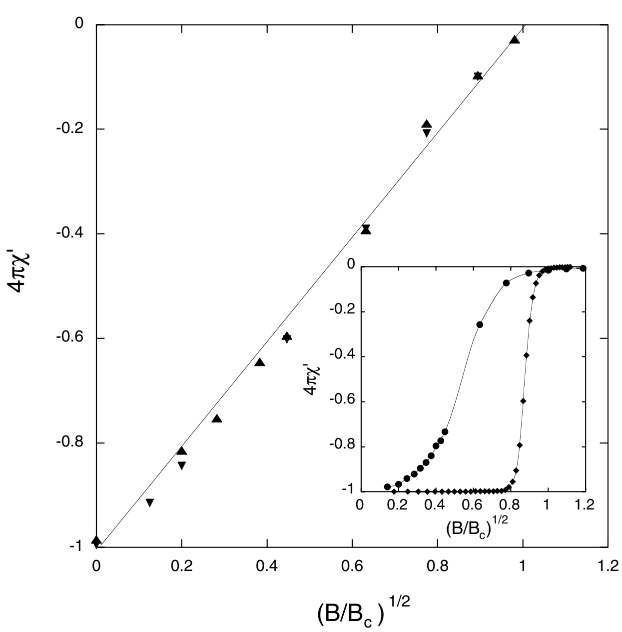

FIG. 4. Square-root field dependence of the susceptibility of two $CePt_3Si$ samples at 70 mK. The line is provided as a guide to the eye. Inset: the circles represent the 10 K susceptibility of the noncentrosymmetric $A15$ superconductor V_3Si. The diamonds are data from a Pb sample alloyed with 5% Sn in order to make it type II. Note that neither of these latter systems obey a square-root scaling law.

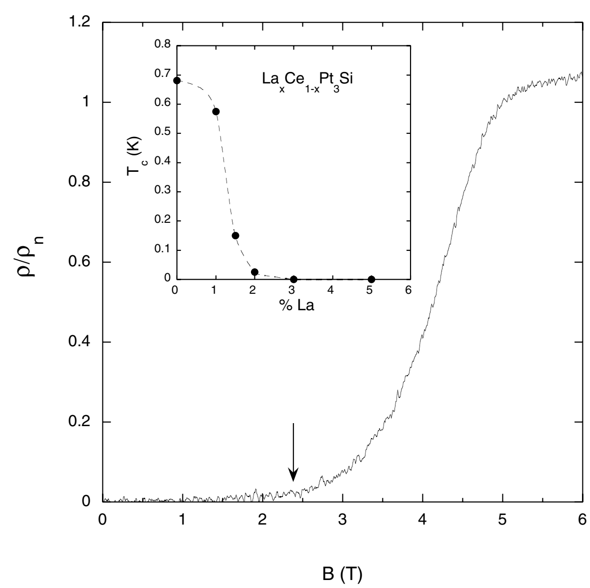

FIG. 5. Transport critical field transition in $CePt_3Si$ at $T = 70$ mK. The arrow depicts the field above which there is no longer a diamagnetic response in the susceptibility. Note that this is also approximately the field at which the onset of resistance occurs. Inset: transition temperature of $Ce_{1-x}La_xPt_3Si$ as a function of x.

$CePt_3Si$ can also be obtained via chemical substitution studies of La for Ce. $LaPt_3Si$ is isotypic to $CePt_3Si$ but is not magnetic and does not superconduct. By substituting nonmagnetic La for Ce one should be able to disorder, and perhaps suppress, the Néel phase without significantly changing the crystal structure. We have, in fact, studied several samples within the series $Ce_{1-x}La_xPt_3Si$, where x varied from 0% to 5%. Correct stoichiometry was confirmed via x-ray analysis. Interestingly, quite small concentrations of La, ~2%, almost completely suppress the superconducting transition temperature; see the inset of Fig. 5. The concurrent effect on the Néel transition is not completely known, but ~2% La is enough to decrease T_N below the base temperature of the PPMS magnetometer, 1.8 K. The extraordinary sensitivity of T_c to La doping is comparable to what one would expect for magnetic impurity doping in a conventional superconductor. A similar suppression of T_c occurs in La doping of the 1-1-5's, but the transition temperature of $Ce_{1-x}La_xCoIn_5$, for instance, is an order of magnitude less sensitive to x than what is shown in Fig. 5 [19]. We assume that the attenuation of T_c is a pair breaking effect, though, of course, La is nonmagnetic. This would suggest that the pairing wave function is unconventional in $CePt_3Si$, and that the enhancement of spin-orbit scattering in this material is producing a pronounced sensitivity to impurities.

In conclusion, the low-temperature susceptibility of superconducting $CePt_3Si$ exhibits a nonhysteretic, square-root power-law field dependence. We believe that these two characteristics of the diamagnetic response are consistent with an unconventional superconducting condensate and a nodal gap structure, in particular. This conjecture is supported by the observed fragility of the superconducting phase to La substitution of Ce. At this point, however, the relative roles of antiferromagnetism, spin-orbit scattering, and the noncentrosymmetric crystal structure in pair formation, and the subsequent expression of a macroscopic quantum state in $CePt_3Si$ are unclear.

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[1] S. Saxena *et al.*, Nature (London) **406**, 587 (2000).
[2] C. Pfleiderer *et al.*, Nature (London) **412**, 58 (2001).
[3] N. Bernhoeft *et al.*, Phys. Rev. Lett. **81**, 4244 (1998); M. Jourdan, M. Huth, and H. Adrian, Nature (London) **398**, 47 (1999).
[4] H. Hegger *et al.*, Phys. Rev. Lett. **84**, 4986 (2000); C. Petrovic *et al.*, Europhys. Lett. **53**, 354 (2001); C. Petrovic *et al.*, J. Phys. Condens. Matter **13**, L337 (2001).
[5] S. Kawasaki *et al.*, Phys. Rev. Lett. **91**, 137001 (2003).
[6] E. Bauer *et al.*, Phys. Rev. Lett. **92**, 027003 (2004).
[7] L. P. Gorkov and E. I. Rashba, Phys. Rev. Lett. **87**, 037004 (2001); S. K. Yip, Phys. Rev. B **65**, 144508 (2002).
[8] V. E. Edelstein, Phys. Rev. Lett. **75**, 2004 (1995); K. V. Samokhin, Phys. Rev. Lett. **94**, 027004 (2005).
[9] P. A. Frigeri, D. F. Agterberg, A. Koga, and M. Sigrist, Phys. Rev. Lett. **92**, 097001 (2004); V. P. Mineev, cond-mat/0405672 v1.
[10] N. Metoki *et al.*, J. Phys. Condens. Matter **16**, L29 (2004).
[11] S. Sachdev, *Quantum Phase Transitions* (Cambridge University Press, Cambridge, 1999).
[12] A. M. Clogston, Phys. Rev. Lett. **9**, 266 (1962); B. S. Chandrasekhar, Appl. Phys. Lett. **1**, 7 (1962).
[13] M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1996).
[14] S. Tewari, D. Belitz, T. R. Kirkpatrick, and J. Toner, Phys. Rev. Lett. **93**, 177002 (2004); L. Radzihovsky, E. M. Ettouhami, K. Saunders, and J. Toner, Phys. Rev. Lett. **87**, 027001 (2001).
[15] G. E. Volovik, JETP Lett. **58**, 469 (1993).
[16] P. W. Anderson and E. I. Blount, Phys. Rev. Lett. **14**, 217 (1965).
[17] P. A. Frigeri, D. F. Agterberg, and M. Sigrist, cond-mat/0405179 v1; M. Yogi *et al.*, Phys. Rev. Lett. **93**, 027003 (2004).
[18] I. Vekhter, P. J. Hirschfeld, and E. J. Nicol, Phys. Rev. B **64**, 064513 (2001).
[19] C. Petrovic, L. Bud'ko, V. G. Kogan, and P. C. Canfield, Phys. Rev. B **66**, 054534 (2002).