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Unusual behaviours and impurity effects in the noncentrosymmetric superconductor $CePt_3Si$

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Abstract. We report a study in which the effect of defects/impurities, growth process, off-stoichiometry and presence of impurity phases on the superconducting properties of noncentrosymmetric $CePt_3Si$ is analysed by means of the temperature dependence of the magnetic penetration depth. We found that the linear low-temperature response of the penetration depth—indicative of line nodes in this material—is robust regarding sample quality, in contrast to what is observed in unconventional centrosymmetric superconductors with line nodes. We discuss evidence suggesting that the broadness of the superconducting transition may be intrinsic, though not implying the existence of a second superconducting transition. The superconducting transition temperature systematically occurs at about 0.75 K in our measurements, in agreement with resistivity and ac magnetic susceptibility data but in conflict with specific heat, thermal conductivity and NMR data in which T_c is about 0.5 K. Random defects do not change the linear low-temperature dependence of the penetration depth

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in the heavy-fermion $CePt_3Si$ with line nodes, as they do in unconventional centrosymmetric superconductors with line nodes.

Contents

1. Introduction

In general, the superconducting Bardeen–Cooper–Schrieffer (BCS) ground state is formed by Cooper pairs with zero total angular momentum. The electronic states are fourfold degenerate: $|\mathbf{k}\uparrow\rangle$, $|-\mathbf{k}\uparrow\rangle$, $|\mathbf{k}\downarrow\rangle$ and $|-\mathbf{k}\downarrow\rangle$ have the same energy $\epsilon(\mathbf{k})$. The states with opposite momenta and opposite spins are transformed into one another under the time-reversal operation $\hat{K}|\mathbf{k}\uparrow\rangle = |-\mathbf{k}\downarrow\rangle$, and the states with opposite momenta are transformed into one another under the inversion operation $\hat{I}|\mathbf{k}\downarrow\rangle = |-\mathbf{k}\downarrow\rangle$. The four degenerate states are a consequence of space and time inversion symmetries. Parity symmetry is irrelevant for spin-singlet pairing, but is essential for spin-triplet pairing. Time-reversal symmetry is required for spin-singlet configuration, but is unimportant for spin-triplet states [1, 2].

All conventional superconductors (s-wave pairing states) are examples of systems invariant under parity and time-reversal symmetries. Some superconductors, like UPt_3 and Sr_2RuO_4, violate time-reversal symmetry and their Cooper pairs form spin-triplet states. The heavy-fermion $CePt_3Si$ is the classic example of a superconductor without inversion symmetry. Parity is not a good quantum number in noncentrosymmetric superconductors; therefore it is not possible to classify their pairing states as pure even-parity spin-singlet or pure odd-parity spin-triplet. Rather, parity mixing is expected. Moreover, the lack of inversion symmetry causes the appearance of an antisymmetric spin–orbit coupling (ASOC) that lifts the spin degeneracy existing in parity-conserving superconducting systems by originating two bands with different spin structures. From this it follows that superconductors without inversion symmetry should show intriguing properties, as indeed is the case for $CePt_3Si$.

Among the striking behaviour of $CePt_3Si$ ($T_c = 0.75$ K) [3], one finds: (a) a small peak just below the superconducting transition in the NMR $1/T_1T$ data [4], characteristic of spin-singlet superconductors with a nodeless energy gap; (b) absence of the Pauli paramagnetic limiting field [3], indicative of spin-triplet pairing which is forbidden by the lack of inversion symmetry; and (c) low-temperature power-law behaviour in the magnetic penetration depth [5] and the thermal conductivity [6], expected for energy gaps with line nodes. However, these conflicting findings have been satisfactorily explained by a model based on the splitting of the spin degenerate band produced by the ASOC [7]–[9].

The superconducting phase of $CePt_3Si$ appears to be very complex [10, 11], and several intriguing issues remain to be elucidated: (a) the broad transition in the electrical resistivity of a clean sample [12], (b) the broad transition and a weak second drop at about 0.5 K in penetration depth [5], (c) a second peak around 0.5 K in specific heat [13, 14] and ac magnetic susceptibility [15, 16] and (d) the occurrence of the superconducting transition at a rather low temperature near 0.5 K in thermal conductivity and some specific heat and NMR data [6], [17]–[19]. The broad transition and second peak have been attributed to the presence of antiferromagnetic impurity phases in the samples [14] and to deviations from the 1 : 3 : 1 nominal stoichiometry and/or structural defects [20, 21]. The second peak has also been interpreted as an indication of a second superconducting transition [13, 15]. The superconducting transition has even been suggested to actually occur at about 0.5 K instead of at 0.75 K [21, 22].

The unusual/conflicting results discussed above seem somewhat sample dependent. However, two of these results have been found in different types of samples: the small peak below the critical temperature in NMR $1/T_1T$ data [4, 19] and the absence of a paramagnetic limiting field [3, 12, 13, 18, 22]. Thus, both properties appear to be intrinsic. Are the low-temperature power laws indicating line nodes affected by sample differences? Which of the puzzling results are intrinsic? These questions need to be addressed in order to understand the physics of $CePt_3Si$ and, in consequence, to have a more complete picture of superconductors without inversion symmetry.

In order to shed light on all these issues we studied the temperature dependence of the magnetic penetration depth of $CePt_3Si$ single crystals in terms of structural defects, impurities and off-stoichiometry. The samples were grown by different groups using different techniques, which allows us to get deep into the sample-dependent problem. The results are compared with those previously obtained in a high-quality polycrystalline sample [5]. The present study suggests that the intrinsic properties of $CePt_3Si$ are more affected by structural defects than by slight off-stoichiometry or the presence of secondary phases. At low temperatures, however, a linear dependence of the penetration depth is observed for all single crystals.

2. Experiment details

We measured four single crystals, labelled A-1, A-2, B-1 and B-2. Crystals A-1 and A-2 were grown from polycrystalline $CePt_3Si$ samples using a mirror furnace. Crystals B-1 and B-2 were prepared by the Bridgman method, in which the growth is extremely slow and the defect density is very low. It is worth mentioning here that conventional annealing, which uses an evacuated quartz tube, is not very effective for Bridgman-grown crystals and is sometimes even harmful. The superconducting critical temperature of the crystals, given in table 1, is defined as the onset of the diamagnetic behaviour in the penetration depth data. All samples had plate-like shapes with dimensions of about $0.5 \times 0.5 \times 0.4\,mm^3$.

Backscattered electron images and both energy-dispersive x-ray (EDX) and wavelength-dispersive x-ray (WDX) spectra of the crystals were obtained with a JEOL Superprobe 8900R electron probe microanalyzer. EDX spectra were used for qualitative analyses. The quantitative analyses of the WDX spectra were performed with the software package CITZAF, by using highly pure Ce, Pt and Si standards and Bastin's phi–rho–z correction method [23]. The stoichiometric formulae of the crystals are shown in table 1. Penetration depth measurements were performed by utilizing a 12 MHz tunnel diode oscillator in a dilution refrigerator running down to the lowest temperature of 40 mK (for details see [5]).

Table 1. Characteristics of the samples used in this work. Single crystals grown by the mirror furnace method are labelled A-n and those grown by the Bridgman technique are labelled B-n.

Sample	T_c (K)	Host phase	Second phase
A-1	0.75	$Ce_{1.14}Pt_3Si_{0.58}$	Yes (rich in Si)
A-2	0.73	$Ce_{1.61}Pt_3Si_{0.91}$	No
B-1	0.79	$Ce_{0.99}Pt_3Si_{1.14}$	Yes (rich in Ce)
B-2	0.79	$Ce_{1.04}Pt_3Si_{1.07}$	No

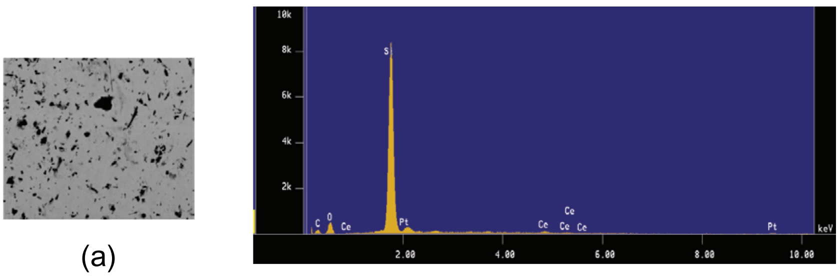

Figure 1. (a) Backscattered electron image of a typical area of the single crystal A-1. Horizontal side: 80 μm. The dark areas correspond to impurity phases. (b) Representative EDX spectrum of the dark areas rich in Si.

3. Results

Single crystals A-1 and B-1 had secondary impurity phases that are seen as dark spots in the backscattered electron images of typical sectional areas displayed in figures 1(a) and 2(a). The EDX spectra of the most abundant impurity phases (dark zones) are shown in figures 1(b) and 2(b). The impurity phases of A-1 (B-1) were rich in Si (Ce), and the host phases of both crystals were off-stoichiometry (see the third column of table 1). Single crystals A-2 and B-2 had pure phases, as can be deduced from the very homogeneous backscattered electron images depicted in figures 3(a) and (b). The single crystal B-2 had an almost nominal stoichiometry of 1 : 3 : 1. The EDX spectrum of crystal B-2 is shown in figure 3(c). It is interesting to note that second impurity phases appeared in samples grown by both the mirror furnace and the Bridgman method.

The effect of growth process, defects, presence of impurity phases and off-stoichiometry on the superconducting properties of $CePt_3Si$ is analysed by means of the temperature-dependent magnetic penetration depth. Figure 4 shows the normalized in-plane penetration depth $\Delta\lambda_{\|}(T)/\Delta\lambda_0$ versus T of all four single crystals of $CePt_3Si$ and, for comparison, $\Delta\lambda(T)/\Delta\lambda_0$ of the polycrystalline sample reported in [5]. Here, $\Delta\lambda_0$ is the total penetration depth shift. Two features are noticeable in this figure: (i) all onset temperatures are about

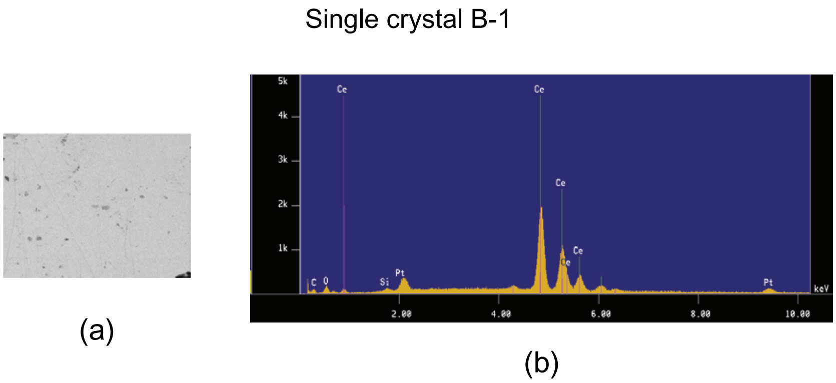

Figure 2. (a) Backscattered electron image of a typical area of the single crystal B-1. Horizontal side: 240 μm. The dark areas correspond to impurity phases. (b) Representative EDX spectrum of the dark areas rich in Ce.

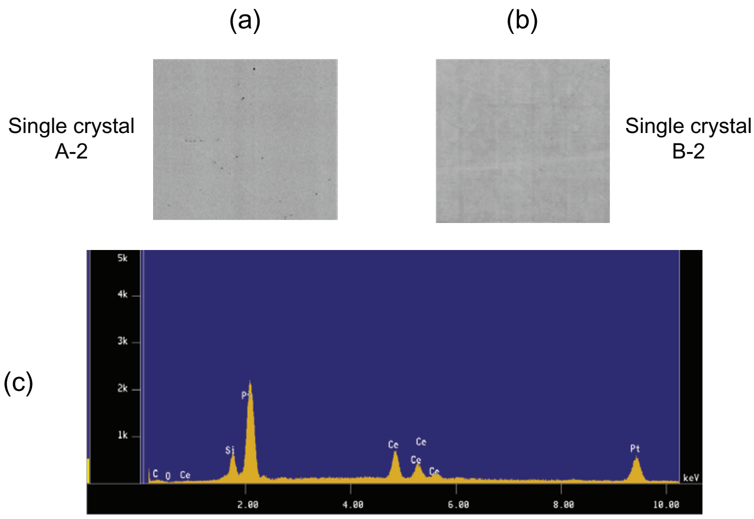

Figure 3. Backscattered electron images of typical areas of single crystals (a) A-2 and (b) B-2. Horizontal side: 240 μm. No dark areas are observed in these images, implying single-phase crystals. (c) EDX spectrum of single crystal B-2.

0.75 K (see table 1) and (ii) the superconducting transitions are broad. The lower transition temperatures and much broader transitions in samples A-1 and A-2, grown by the mirror furnace technique, are most likely due to the presence of structural defects and/or random impurities since crystal A-2 had no impurity phases. Crystals B-1 and B-2 present higher critical temperatures and less broad transitions than the high-quality annealed polycrystalline sample studied in [5] (see table 1). The penetration depth data of crystals B-1 and B-2 are similar, even though crystal B-1 had second impurity phases.

To study the effect of sample quality on the line nodes of $CePt_3Si$ [5, 6], we depict in figure 5 the low-temperature region of $\Delta\lambda_{\|}(T)/\Delta\lambda_0$ for our four single crystals. The linear temperature dependence of the penetration depth—indicating line nodes in the energy gap—found in an annealed polycrystalline sample [5] is also detected in single crystals. The

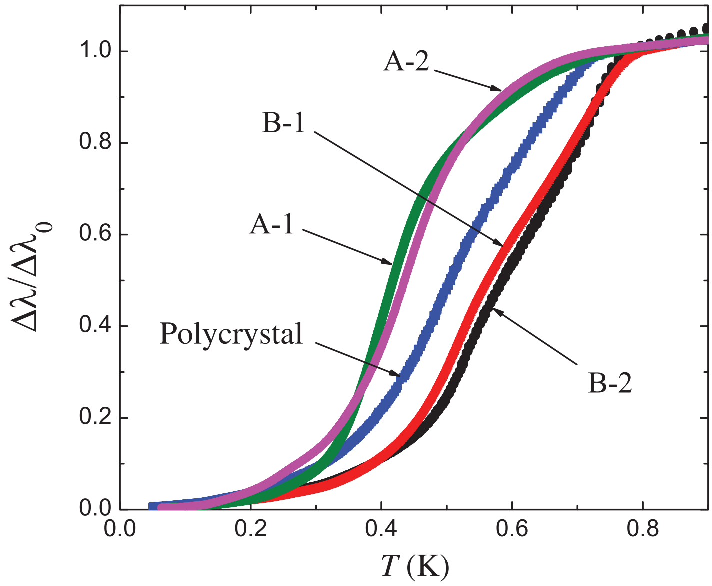

Figure 4. Normalized in-plane penetration depth $\Delta\lambda_{\parallel}(T)/\Delta\lambda_0$ versus T of our four single crystals of $CePt_3Si$. For comparison, data of a polycrystalline sample (from [5]) are also shown.

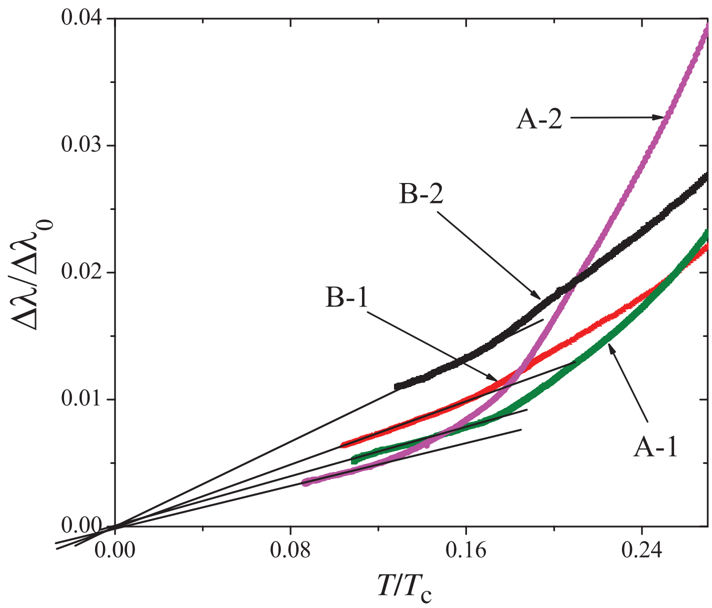

Figure 5. Low-temperature region of normalized in-plane penetration depth $\Delta\lambda_{\parallel}(T)/\Delta\lambda_0$ versus T/T_c of our four single crystals of $CePt_3Si$. A linear low-temperature behaviour is observed in all crystals, as it was found previously in a polycrystalline sample [5].

outstanding observation in figure 5, however, is that sample quality does not affect such a linear temperature behaviour. The line nodes of $CePt_3Si$ are robust regarding disorder and the mechanism causing the broadening of the superconducting transition.

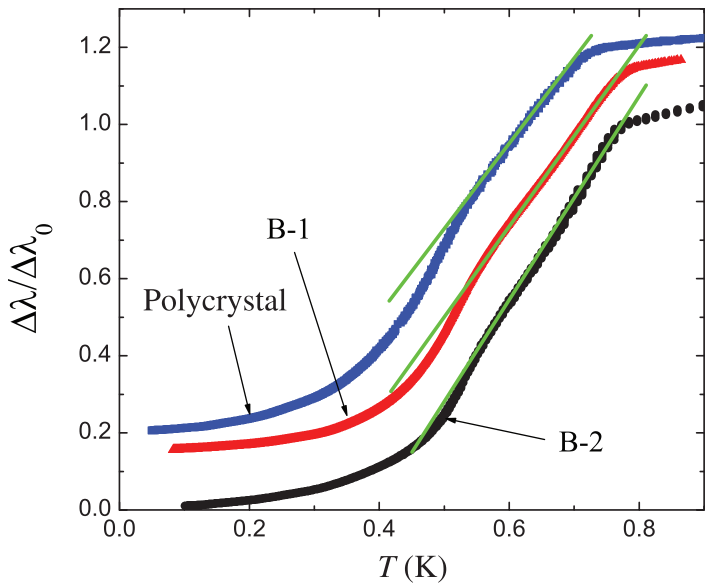

Figure 6. Normalized in-plane penetration depth data of crystals B-1 and B-2, which have the sharper superconducting transitions. Data for a polycrystalline sample (from [5]) are also shown. Data for the polycrystalline and B-1 samples have been shifted for clarity.

4. Discussion

The fact that the superconducting transition of $CePt_3Si$ as measured by penetration depth is wide, independent of the growth procedure, absence of impurity phases and stoichiometry, agrees with all previous results obtained by other techniques in all kinds of samples, varying from unannealed/quenched/annealed polycrystalline samples to annealed single crystals. The sharpest transitions, with a width of about 0.15 K, have been seen in annealed polycrystalline samples [21] and annealed single crystals grown by the Bridgman technique [18]. For the latter ones it was even possible to observe de Haas–van Alphen oscillations, a clear indication of the low-disorder character of these samples with a mean free path $l = 1200–2700$ Å much larger than the coherence length $\xi(0) \sim 100$ Å. We remark that a width of 0.15 K is quite large considering the transition temperature of 0.75 K. From our results, we believe that impurity phases and off-stoichiometry do not play significant roles in the broadness of the transition.

Is the transition width related to the occurrence of an inflection point at about 0.5 K in the penetration depth and a second peak in some specific heat data? The answer seems to be 'No'. In specific heat measurements the transition is always broad, independent of whether it occurs at 0.7 K [3, 13, 14, 21], in which case sometimes two peaks appear, or at 0.5 K [17, 18, 21], in which case only one peak is always observed. To verify what is seen in penetration depth measurements, we plot again in figure 6 the penetration depth data with the sharper superconducting transitions, where the data for the polycrystalline and B-1 samples have been shifted for clarity. In our best sample (B-2), the inflection point is barely observed, yet the transition is wide. We notice that the second less strong drop in the penetration depth shows up independently of the existence of the second impurity phases. It is possible that in a slightly better sample, with fewer defects, the second drop withers, whereas the transition remains broad. We argue here that the second anomaly in either specific heat or penetration depth of $CePt_3Si$ does not have an intrinsic superconducting origin. A second superconducting transition, suggested in previous reports [13, 15], is discarded. The transition broadness could be an effect of lack of inversion symmetry; however, it has not been observed in other noncentrosymmetric superconductors.

Our penetration depth data display the superconducting transition at about 0.75 K. Is this the true superconducting transition temperature of $CePt_3Si$ or is it 0.5 K? We argue that sample quality does not make a difference in this issue. For example, for the same single crystal, T_c was 0.5 K when probed by thermodynamic properties like specific heat and thermal conductivity, but was 0.75 K when measured by resistivity [6, 18]. In general, T_c of $CePt_3Si$ depends on how it is measured; that is, it depends on the measuring technique. The transition takes place at about 0.5 K in thermodynamic measurements like specific heat [17, 18, 21] and thermal conductivity [6] and in magnetic probes like NMR $1/TT_1$ [19]. It occurs at 0.75 K in inductive methods like penetration depth and ac susceptibility [15, 20, 22, 24] and in resistivity [6, 18]. An odd result comes from a recent ac magnetic susceptibility measurement that found a broad transition at 0.5 K [25]. Interestingly, within each set of measuring techniques giving different T_c's, the probes have no physical connection with one another. This makes the case of $CePt_3Si$ different from that of the heavy-fermion superconductor $CeIrIn_5$, in which resistivity measurements show the superconducting transition at about 1.2 K, whereas thermodynamic and magnetic properties display it at 0.4 K [26]. At present, the origin of this inconsistency in $CePt_3Si$ is not clear to us.

To discuss the low-temperature response of the penetration depth, we consider that the absence of inversion symmetry in a crystal structure causes the appearance of an antisymmetric potential gradient ∇V that leads to an ASOC $\alpha(\mathbf{k}\times\nabla V)\cdot\hat{\sigma}$. Here $\mathbf{k}$ is the electron momentum and α a coupling constant. In superconductors, a strong ASOC lifts the spin degeneracy existing in parity-conserving systems by originating two bands with different spin structures and with energy gaps [7]

$$\Delta_{\pm}(\mathbf{k}) = \psi \pm t|\mathbf{g}(\mathbf{k})|. \tag{1}$$

Here ψ and $t|\mathbf{g}(\mathbf{k})|$ are the spin-singlet and spin-triplet components, respectively, and $\mathbf{g}(\mathbf{k})$ is a dimensionless vector $[\mathbf{g}(-\mathbf{k}) = -\mathbf{g}(\mathbf{k})]$ parallel to the vector $\mathbf{d}(\mathbf{k})$ of the spin-triplet order parameter. In this spin-split band model the order parameter is then a mixture of spin-singlet and spin-triplet states, and when the spin–orbit band splitting E_{so} is much larger than the superconducting energy scale $k_B T_c$ the system can be essentially regarded as a two-gap superconductor with gaps that open at T_c. In $CePt_3Si$ it is assumed [7] that the gaps in equation (1) have an isotropic s-wave spin-singlet component and a p-wave spin-triplet component with $\mathbf{g}(\mathbf{k}) \propto (k_y, -k_x, 0)$. This spin-triplet component has point nodes.

Line or point nodes in the energy gap structure are natural and are required by symmetry in some unconventional centrosymmetric superconductors, but their existence is not obvious in parity-violating superconductors. The line nodes in $CePt_3Si$, however, can be confidently explained by using the spin-split band model [9]. They turn out to be accidental—not imposed by symmetry—and appear in the gap $\Delta_-(\mathbf{k}) = \psi - t|\mathbf{g}(\mathbf{k})|$ when $(\psi/t) < 1$. For $CePt_3Si$, in particular, $(\psi/t) \sim 0.6$, and the energy gap $\Delta_-(\mathbf{k})$ has the three-dimensional (3D) polar representation shown in figure 7.

The broad penetration depth response below T_c leads to a suppression of the superfluid density in the high-temperature region. To expound such a suppression and the line nodes, the

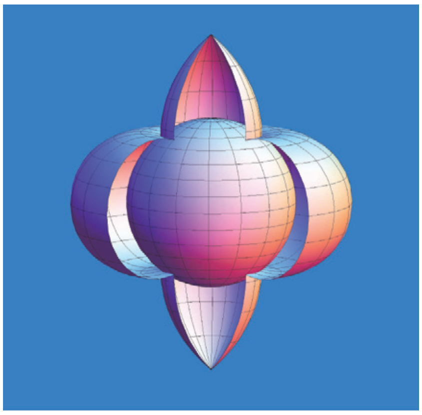

Figure 7. Three-dimensional polar plot of the gap function $\Delta_-(\mathbf{k})$ in $CePt_3Si$.

spin-split band model would require that the band with gap $\Delta_-(\mathbf{k})$ has a normalized density of states of about 0.9 [9], whereas energy band calculations indicate that the difference in the density of states of the spin-split bands is of the order of 0.3 [27]. Thus, at present, the broadness of the superconducting transition cannot be accounted for in this model (see [9] for details).

We will now discuss the effect of impurities and other defects that may exist in our crystals on the superconducting behaviour of noncentrosymmetric $CePt_3Si$. As we have mentioned above, the transition broadness and lower critical temperatures in samples A-1 and A-2 are probably related to defects and/or random impurities. We note that low defect and/or nonmagnetic impurity concentrations affect the low-temperature properties of unconventional one-band superconductors with inversion symmetry. In the case of symmetry-required line nodes of a one-component order parameter, it was proposed that in the unitary scattering limit there is a crossover temperature $T^* \sim (\Gamma\Delta_0)^{1/2}$ from a high-temperature linear to a low-temperature quadratic behaviour of the penetration depth [28]. Γ is a scattering rate that depends on the impurity concentration. For strong scattering, a relatively small concentration of defects would cause a high value of T^* without significantly depressing T_c. This is indeed observed in, for example, unconventional d-wave high-T_c superconductors [29]. In noncentrosymmetric $CePt_3Si$ with line nodes, such a behaviour is not seen. If one assumes that the optimum onset temperature in $CePt_3Si$ is that of the B crystals ($T_{c0} = 0.79$ K) and that the lower onset temperature of crystal A-2 ($T_c = 0.73$ K) is due to the presence of defects/impurities, one has for the latter sample a relative T_c suppression $(T_{c0} - T_c)/T_{c0}$ of about 8% and a $T^* \sim 0.38T_c$ (using $\Delta_0/T_c = 2.15$ for a line-node gap). For crystal A-1, the relative T_c suppression is 5% and $T^* \sim 0.31T_c$. According to these estimations, in crystals A-1 and A-2 the penetration depth should follow a T^2 behaviour below about $0.3T_c$, which is not observed in the data shown in figure 5. The penetration depth instead becomes linear below $T \approx 0.16T_c$ in A-1 and $T \approx 0.12T_c$ in A-2.

From the above analysis, it seems that in parity-violating $CePt_3Si$ with line nodes, defects or nonmagnetic impurities do not perturb the low-temperature penetration depth in the same manner as in unconventional centrosymmetric superconductors. We notice here the observation of weak pair-breaking effects of nonmagnetic-ion substitutions on Pt and Si sites [30, 31], where rare-ion concentrations as high as 6 and 10% do not destroy superconductivity. This apparent insensitivity to disorder has resemblances to a conventional superconducting phase and suggests that the superconducting order parameter of $CePt_3Si$ is not an even basis function that transforms according to the nontrivial 1D irreducible representations of C_{4v} (A_2, B_1 and B_2). Recent theoretical works [32, 33] considered impurity effects on the critical temperature and the density of states [34] of superconductors without inversion symmetry. It was found that for some particular cases impurity scattering leads to a functional form of T_c that, up to a prefactor, is the same as the one for unconventional superconductors with inversion symmetry: $\ln(T_c/T_{c0}) = \alpha[\Psi(\frac{1}{2}) - \Psi(\frac{1}{2} - \frac{\Gamma}{2\pi T_c})]$. For $CePt_3Si$ $\alpha \sim 0.9$, therefore there is not much change in the estimations of T^* carried out in the paragraph above. In the analysis of the local density of states it was concluded that a single nonmagnetic impurity-induced resonant state near the Fermi energy is likely possible in noncentrosymmetric superconductors [34]. This implies that defects or nonmagnetic impurities would perturb the low-T response of thermodynamic variables and the penetration depth, a disturbance that is not observed in our low-temperature $\lambda(T)$ data.

5. Conclusions

We reported on magnetic penetration depth measurements on several single crystals of $CePt_3Si$ grown by different techniques. We discussed the effects of the growth processes, presence of impurity phases, off-stoichiometry and defects on the superconducting phase of this compound. We found the following:

1. The presence of impurity phases and off-stoichiometry have a relatively low influence on the broadness of the superconducting transition. On the other hand, the growth process, related to the disorder in the samples, has a higher impact on the transition widening. The superconducting transition in $CePt_3Si$ may be intrinsically wide.
2. The anomaly at about 0.5 K, observed in several superconducting properties, appears to fade out in the penetration depth data as the transition gets sharper.
3. In penetration depth measurements, the superconducting transition occurs at about 0.75 K, as also found in resistivity and ac magnetic susceptibility measurements. This critical temperature contrasts the one at 0.5 K detected in specific heat, thermal conductivity and NMR data.
4. Defects and/or impurities do not change the linear low-temperature dependence of the penetration depth of noncentrosymmetric $CePt_3Si$ with line nodes, as they do in other unconventional superconductors with line nodes in the gap.

Acknowledgments

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