Evidence of nodal gap structure in the noncentrosymmetric superconductor Y_2C_3

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The magnetic penetration depth $\lambda(T)$ and the upper critical field $\mu_0 H_{c2}(T_c)$ of the non-centrosymmetric superconductor Y_2C_3 have been measured using a tunnel-diode-based resonant oscillation technique. We found that the penetration depth $\lambda(T)$ and its corresponding superfluid density $\rho_s(T)$ show linear temperature dependence at very low temperatures ($T \ll T_c$), indicating the existence of line nodes in the superconducting energy gap. Moreover, the upper critical field $\mu_0 H_{c2}(T_c)$ presents a weak upturn at low temperatures with a rather high value of $\mu_0 H_{c2}(0) \simeq 29$ T, which slightly exceeds the weak-coupling Pauli limit. We discuss the possible origins for these nontrivial superconducting properties, and argue that the nodal gap structure in Y_2C_3 is likely attributed to the absence of inversion symmetry, which allows the admixture of spin-singlet and spin-triplet pairing states.

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I. INTRODUCTION

The symmetries of a superconductor, e.g., time-reversal and spatial inversion symmetries, may impose important constraints on the pairing states. Among the previously investigated superconductors, most possess an inversion center in their crystal structures, in which the Cooper pairs are either in an even-parity spin-singlet or odd-parity spin-triplet pairing state, according to the Pauli principle and parity conservation.[1,2] However, the tie between spatial symmetry and the Cooper-pair spins might be violated in a superconductor lacking inversion symmetry.[3–7] In these materials, an asymmetric potential gradient yields an antisymmetric spin-orbit coupling (ASOC), which splits the Fermi surface into two spin-ordered subsurfaces, with pairing allowed both across one subsurface and between the two. ASOC splits the degeneracy of conduction electrons, and allows the admixture of spin-singlet and spin-triplet pairing states within the same orbital channel. Various exotic electromagnetic properties may arise in the parity-violated materials.[3–7]

Until now, only a very few noncentrosymmetric (NCS) superconductors have been investigated. In the heavy fermion NCS superconductors, e.g., $CePt_3Si$ and $CeMSi_3$ ($M =$ Rh,Ir), the upper critical field is greatly enhanced in comparison with that of other heavy fermion materials,[8–10] exceeding the Pauli limit in a weak-coupling BCS model. Measurements of thermodynamic properties in these compounds indicated the existence of line nodes in the superconducting gap, even though a weak Hebel-Slichter coherence peak is observed below T_c.[11–15] These unconventional features were interpreted in terms of a Rashba-type spin-orbit coupling model arising from the absence of inversion symmetry.[5–7] However, the strong electronic correlations and the closeness to a magnetic instability might complicate the analysis of the ASOC effect on superconductivity. Recent work on $Li_2(Pd_{1-x}Pt_x)_3B$ systems demonstrated that spin-singlet and spin-triplet order parameters can add constructively and destructively, leading to two different gap functions and the possible occurrence of accidental nodes in the destructive gap.[16–19] Mo_3Al_2C seems to present another example of a weakly correlated NCS superconductor,[20] showing evidence of an unconventional pairing state attributed to the lack of inversion symmetry. Furthermore, multigap superconductivity was also suggested for La_2C_3 (Ref. 21) and $Mg_{10}Ir_{19}B_{16}$.[22] To look into the effect of ASOC on superconductivity, a search for unconventional superconductivity in weakly correlated noncentrosymmetric compounds remains highly desirable.

The nonoxide transition metal sesquicarbides M_2C_3 ($M =$ La,Y), which crystallize in a cubic Pu_2C_3-type structure (space group $I\bar{4}3d$), present another family of NCS superconductors with a relatively high T_c ($T_c \sim 18$ K).[23–25] Resembling the $Li_2(Pd_{1-x}Pt_x)_3B$ system[16–19] and Mo_3Al_2C,[20] M_2C_3 shows no evidence of strong electronic correlations and/or magnetic fluctuations. Thus it might provide another example in which to study the effect of ASOC on superconductivity. Measurements of nuclear magnetic resonance[26] (NMR) and muon spin relaxation[27] (μSR) indicate a complex gap structure in Y_2C_3 which cannot be described in terms of a simple *s*-wave or *d*-wave gap function, but can be qualitatively fitted with a two-gap model at temperatures near T_c. While there is some variation among the weights reported, the fits near T_c all give a dominant large gap of $2\Delta_1/k_B T_c \approx 5$ and a small gap of $2\Delta_2/k_B T_c \approx 1.1$–2. On the other hand, the Knight shift in NMR decreases to approximately 2/3 of its normal-state value, and the temperature dependence of $1/T_1$ deviates from the predictions of a conventional BCS model.[26] Recently, we noticed that Y_2C_3 shows a high upper critical field [$\mu_0 H_{c2}(0) \simeq 29$ T], which might indicate the importance of the ASOC effect in this compound.[28]

In order to elucidate further the pairing state in Y_2C_3, here we report an accurate measurement of the temperature dependence of the resonant frequency shift $\Delta f(T)$ down to 90 mK using a tunnel-diode- (TDO-)based resonant oscillator. The penetration depth $\lambda(T)$ and the corresponding superfluid density $\rho_s(T)$ derived from $\Delta f(T)$ can be well interpreted by a two-gap model at temperatures near T_c. Remarkably, however, a linear-type temperature dependence of $\lambda(T)$ and $\rho_s(T)$ is observed at very low temperatures ($T \ll T_c$), providing evidence for the existence of line nodes in the energy gap.

Together with the observation of a high upper critical field [$\mu_0 H_{c2}(0) \simeq 29$ T] in Y_2C_3, we argue that ASOC might give rise to the admixture of spin-singlet and spin-triplet pairing states as previously discussed in $Li_2(Pd_{1-x}Pt_x)_3B$ systems,[16] leading to the appearance of a nodal gap structure at low temperatures.

II. EXPERIMENTAL METHODS

Polycrystalline samples of Y_2C_3 were prepared by an arc-melting method, followed by heat treatments under high-temperature and high-pressure conditions to form the sesquicarbide phase.[25] Powder x-ray diffraction identified the achieved ingots as a single phase. Precise measurements of the resonant frequency shift $\Delta f(T)$ were performed using a TDO-based, self-inductive technique at 21 MHz in a dilution refrigerator down to 90 mK.[29] The change in penetration depth $\Delta\lambda(T)$ is proportional to $\Delta f(T)$, i.e., $\Delta\lambda(T) = G\Delta f(T)$, where the G factor is a constant determined by sample and coil geometries and, therefore, varies from sample to sample. It is noted that the samples of Y_2C_3 investigated here are rather air sensitive and we could not polish the surface to form a regular geometry, which might result in a relatively large uncertainty in the determination of the G factor when following traditional methods as described in Ref. 29. Nevertheless the temperature-dependent behavior of $\Delta\lambda(T)$ remains reliable and unchanged. The upper critical field $\mu_0 H_{c2}(T_c)$ was also determined from a similar TDO-based resonant oscillation method in a pulsed magnetic field.[28]

III. RESULTS AND DISCUSSION

In Fig. 1, we plot the temperature dependence of $\Delta f(T)$ for three samples of Y_2C_3, which were cut either from the same batch (nos. 2-1,2-2) or from a different batch (no. 4-2). One can see that all three samples follow similar behavior. The sharp drop of $\Delta f(T)$ marks a superconducting transition with $T_c \simeq 15$ K. The linear temperature dependence of $\Delta f(T)$ is reproducibly achieved at low temperatures as shown in the insets of Fig. 1, significantly deviating from the exponential behavior of conventional BCS superconductors. Such linear temperature dependence of $\Delta\lambda(T)$ gives strong evidence for the existence of line nodes in the superconducting energy gap of Y_2C_3, remarkably resembling the case of $Li_2(Pd_{1-x}Pt_x)_3B$ ($x \geqslant 0.3$).[16,17] The different absolute values of $\Delta f(T)$ are mainly attributed to the distinct values of the G factor for each sample.

The superfluid density is directly related to the superconducting gap structure on the Fermi surface. In Fig. 2, we plot the superfluid density $\rho_s(T)$ for Y_2C_3 (circles), derived using $\rho_s(T) = \lambda_0^2/\lambda^2(T)$. Here $\lambda(T) = \lambda_0 + \Delta\lambda(T)$, in which $\lambda_0 = 470$ nm is the penetration depth at zero temperature,[27] and the G factor is 0.45 nm/Hz for sample no. 4-2. For comparison, $\rho_s(T)$ obtained from the μSR experiments is also shown in Fig. 2 (see the triangles in the main figure).[27] The agreement between the two measurements is excellent. However, our penetration depth measurements extend $\rho_s(T)$ to much lower temperatures, which provides significant inputs into the superconducting pairing state (see below).

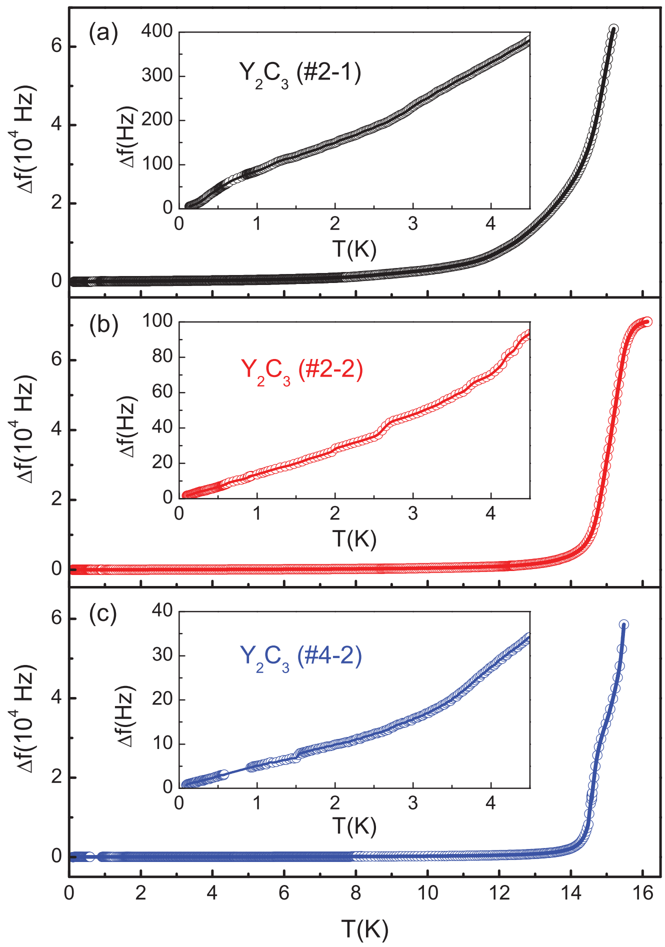

FIG. 1. (Color online) Temperature dependence of the resonant frequency shift $\Delta f(T)$ for various Y_2C_3 samples. The insets plot $\Delta f(T)$ at low temperatures, showing linear-type temperature dependence.

A two-gap scenario, as previously discussed in MgB_2,[30,31] has been proposed to describe the NMR and μSR data for Y_2C_3.[26,27] Here we tried to fit $\rho_s(T)$ with a two-gap BCS model:

$$\rho_s(T) = x\delta\rho_s\left(\Delta_1^0,T\right) + (1-x)\delta\rho_s\left(\Delta_2^0,T\right), \tag{1}$$

where Δ_i^0 ($i = 1$ and 2) are the energy gaps at $T = 0$ and x is the relative weight for Δ_1^0. The gap functions are given by

$$\delta\rho_s(\Delta,T) = \frac{2\rho_s(0)}{k_B T}\int_0^\infty f(\epsilon,T)[1 - f(\epsilon,T)]d\epsilon, \tag{2}$$

where $f(\epsilon,T) = (1 + e^{\sqrt{\epsilon^2+\Delta^2(T)}/k_B T})^{-1}$ is the Fermi distribution function. Here, k_B is the Boltzmann constant and $\Delta_i(T)$ were taken to follow the universal BCS temperature dependence.

Indeed the two-gap model can describe the overall superfluid density $\rho_s(T)$ of Y_2C_3 as shown by the dotted line in Fig. 2, particularly in the high-temperature region near T_c. Such a fitting gives the following parameters: $2\Delta_1^0/k_B T_c = 4.9$, $2\Delta_2^0/k_B T_c = 1.1$, and $x = 0.86$, which are in good agreement with those in NMR[26] and μSR measurements.[27] However, significant deviation of $\rho_s(T)$ from the two-gap model is observed at temperatures below $0.4T_c$ [see inset (a)

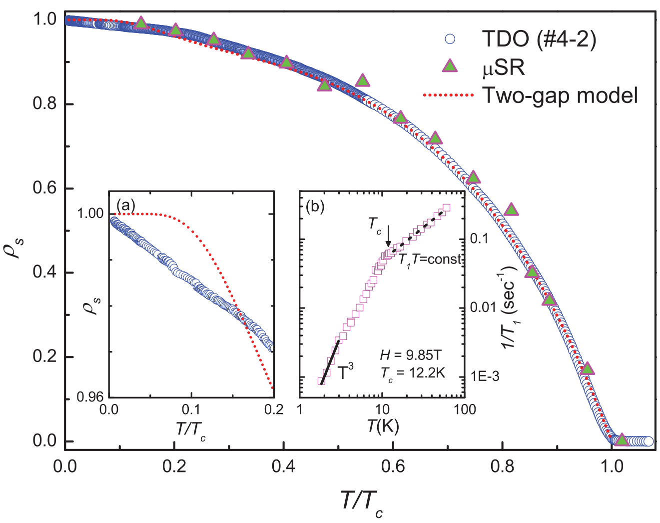

FIG. 2. (Color online) The normalized superfluid density $\rho_s(T)$ versus temperature for Y_2C_3. The circles represent our experimental data derived from the penetration depth $\lambda(T)$ of sample no. 4-2. The filled triangles are digitized from Ref. 26, which were determined from μSR experiments. The dotted line indicates the best fitting using the two-gap model described in the text. Inset (a) shows $\rho_s(T)$ and the fittings in the low-temperature region and (b) shows the temperature dependence of $1/T_1$ for Y_2C_3 (from Ref. 26), respectively.

in Fig. 2]. Our data clearly show $\rho_s(T) \sim T$ at the lowest temperatures, providing strong evidence for the occurrence of line nodes at low temperatures. Such behavior cannot be described by a simple two-gap BCS model. It is noted that our reanalysis of the NMR data reported in Ref. 26 suggests $1/T_1 \sim T^3$ at $T < 3$ K [see inset (b) in Fig. 2], consistent with our TDO measurements and further supporting the existence of line nodes rather than a fully opened gap in Y_2C_3. However, accurate measurements of the muon relaxation rate and/or the nuclear spin-lattice relaxation rate at lower temperatures are needed.

The nodal superconducting gap structure, together with a weak Hebel-Slichter coherence peak,[26] is rarely observed in a simple metallic superconductor which shows no evidence of strong electronic correlations and magnetic fluctuations. As we know, the following three possibilities may lead to a nodal gap structure within the phonon pairing mechanism. First, exotic superconductivity with a nodal gap structure may appear in a multiband system in which the different sheets of Fermi surface are connected via some "necks."[32] However, in this case the superconducting transition is expected to be accompanied by some kind of magnetic order as a result of broken time-reversal symmetry. Indeed, the band structure calculations indicate that the Fermi surface of Y_2C_3 consists of three hole bands and one electron band, arising mainly from the Y-4d and C-2$p\pi^*$ hybridization.[33] The differences in the density of states and Fermi velocities between hole and electron bands might lead to two superconducting gaps opening in different parts of the Fermi surface. However, there is no evidence of any magnetic order in Y_2C_3 and, therefore, one may exclude such a scenario here. Recently, Fujimoto studied the case of a noncentrosymmetric superconductor with a weak ASOC and found that a field-induced pair correlation between the different spin-orbit-split bands might yield a pointlike anisotropic gap;[34] this hardly describes our experimental findings at zero field. As an alternative, we argue that the ASOC effect arising from the broken inversion symmetry might mix the spin-singlet and spin-triplet pairing states, leading to the existence of line nodes in the destructive gap as we previously observed in $Li_2(Pd_{1-x}Pt_x)_3B$ systems.[16,17] In the noncentrosymmetric superconductor Y_2C_3, the band splitting due to ASOC effect is compatible with the superconducting gap,[33] a situation similar to that of $x = 0.3$ in $Li_2(Pd_{1-x}Pt_x)_3B$.[17] In this case, the contribution of the spin-triplet component might be compatible with or even slightly larger than that of the spin-singlet component, resulting in an anisotropic gap with accidental nodes at a small fraction of the Fermi surface which might then lead to a weak linear temperature dependence in the penetration depth $\lambda(T)$ at the lowest temperatures.

The upper critical field $\mu_0 H_{c2}(T_c)$ determined by a similar TDO-based method in a pulsed magnetic field further supports our argument mentioned above. In Fig. 3, the magnetic field dependence of the relative frequency shift $\Delta f(\mu_0 H)$ at selected temperatures is shown for sample no. 2-1. The sudden increase of $\Delta f(\mu_0 H)$ upon decreasing magnetic field marks the superconducting transition. The values of $\mu_0 H_{c2}$ are determined from the onsets of the superconducting transition, at which Δf deviates from its linear field dependence in the normal state (see the arrows in the inset). Obviously, superconductivity is eventually suppressed by applying a magnetic field and vanishes around 29 T.

In Fig. 4 we plot the temperature dependence of $\mu_0 H_{c2}(T_c)$ down to 0.5 K for the samples nos. 2-1 and 2-2, respectively. The following two points can be derived from Fig. 4: First, the upper critical field shows very unusual temperature dependence, $\mu_0 H_{c2}(T_c)$ increasing linearly with decreasing temperature near T_c but showing a weak upturn at low temperatures. Such temperature dependence of $\mu_0 H_{c2}(T_c)$ cannot be described either by the weak coupling Werthamer-Helfand-Hohenberg method[35] or the Ginzburg-Landau theory ($\mu_0 H_{c2}(T) = \mu_0 H_{c2}(0)[1 - (T/T_c)^2]/[1 + (T/T_c)^2]$), as

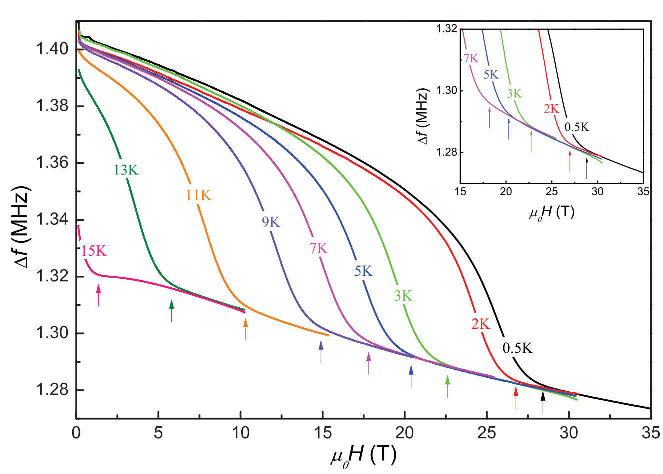

FIG. 3. (Color online) The magnetic field dependence of the frequency shift $\Delta f(\mu_0 H)$ at various temperatures for Y_2C_3 (sample no. 2-1). The inset shows an expanded view of the superconducting transitions at low temperatures. The upper critical field $\mu_0 H_{c2}$ is determined from the superconducting onset as marked by the arrows.

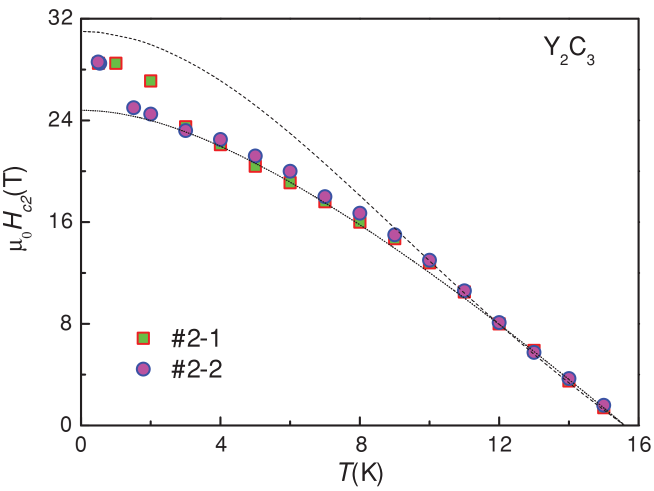

FIG. 4. (Color online) The upper critical field versus temperature for two samples of Y_2C_3: nos. 2-1 and 2-2. The dotted and the dashed lines are fits to the weak-coupling Werthamer-Helfand-Hohenberg (WHH) method and the Ginzburg-Landau (GL) theory, respectively.

shown in Fig. 4. Second, our direct measurements give a rather high upper critical field of $\mu_0 H_{c2}(0) \simeq 29$ T, which is higher than the orbitally limited field $[\mu_0 H_{c2}^{\mathrm{orb}}(0) = -0.69 T_c (d\mu_0 H_{c2}/dT)_{T_c} = 24.5$ T] but is compatible with the paramagnetic limiting field $[\mu_0 H_{c2}^{P}(0) = (1.86\ \mathrm{T/K}) T_c = 28.8$ T] derived for a weak BCS superconductor. Such an enhancement of $\mu_0 H_{c2}(0)$ was previously observed in the noncentrosymmetric superconductors $CePt_3Si$, $CeRhSi_3$, and $CeIrSi_3$,[8–10] presumably attributed to the contribution of the spin-triplet component in the mixed pairing state.[5,7] Therefore, observations of these unusual features in $\mu_0 H_{c2}(T_c)$ might strengthen the importance of the ASOC effect for the superconducting pairing states in Y_2C_3.

IV. CONCLUSION

In summary, we have investigated the penetration depth $\lambda(T)$ and the upper critical field $\mu_0 H_{c2}(T_c)$ of the noncentrosymmetric superconductor Y_2C_3 using a TDO-based resonant oscillator. We found that a two-gap model can well describe the superfluid density $\rho_s(T)$ at temperatures near T_c, but is clearly violated at low temperatures. The frequency shift $\Delta f(T)$ and, therefore, the corresponding penetration depth $\Delta\lambda(T)$ and superfluid density $\rho_s(T)$ show a weak linear temperature dependence at the lowest temperatures, indicative of the existence of line nodes in the superconducting energy gap. Together with the observation of an enhanced upper critical field $[\mu_0 H_{c2}(0) \simeq 29$ T], we argue that these nontrivial superconducting properties might be attributed to the absence of an inversion symmetry in Y_2C_3, in which the ASOC splits the electronic bands, mixing the spin-singlet and spin-triplet pairing states and, therefore, leading to the existence of line nodes. Our findings indicate that Y_2C_3 might present another important example to study the effect of ASOC on superconductivity.

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