Magnetic response of noncentrosymmetric superconductor La$_2$C$_3$:
Effect of double-gap and spin-orbit interaction

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Abstract

The presence of spin-orbit (SO) interaction in a noncentrosymmetric superconductor, La$_2$C$_3$ ($T_c \approx 11$ K) is demonstrated by muon spin rotation ($\mu$SR) in its normal state, where $\mu$SR spectra exhibit field-induced weak depolarization due to van Vleck-like local susceptibility. In the mixed state, muon spin relaxation due to inhomogeneity of internal field ($\sigma_v$) exhibits a field dependence that is characterized by a kink, where $\sigma_v$ (and hence the superfluid density) is more strongly reduced at lower fields. This is perfectly in line with the presence of a secondary energy gap previously inferred from the temperature dependence of $\sigma_v$, and also consistent with the possible influence of asymmetric deformation of the Fermi surface due to the SO interaction.

Key words: superconductivity, spin-orbit interaction, quasiparticle excitation

Multigap superconductivity is interesting in its own right as a manifestation of anisotropic superconductivity. Even within the framework of the BCS mechanism, anisotropy in crystal structure may lead to multi-band structure and associated complex superconducting order parameter because of the possible coupling of electrons to different phonon modes upon formation of the Cooper pairs in the respective energy bands. The discovery of high-$T_c$ superconductivity in magnesium diboride (MgB$_2$)[1] and subsequent struggle for proper understanding of its double-gap nature associated with $\sigma$- and $\pi$-bands brought the multigap superconductivity to the forefront of considerable attention in this field.

Recently, we have shown in a pair of sesquicarbide superconductors $Ln_2C_3$ ($Ln$=La, Y) that the multigap structure would take great variety in its appearance, where, despite a common double-gap structure in the order parameter, the temperature dependence of superfluid density exhibits a remarkable difference between La$_2$C$_3$ and Y$_2$C$_3$ that is understood as resulting from the alteration of interband coupling upon substitution of La for Y.[2]

Meanwhile, sesquicarbides are drawing further attention as a possible stage for exotic superconductivity caused by their noncentrosymmetric crystal structure.[3] The absence of inversion symmetry leads to mixing of parity in the Cooper pairs, making it irrelevant to classify the states in terms of spin-multiplet. It also gives rise to the spin-orbit (SO) interaction that may affect superconductivity as well as the electronic property of normal (paramagnetic) state. In particular, $Ln_2C_3$ may be in a unique situation that the SO interaction is of comparable magnitude to that of superconducting gap,
so that pair correlation between subbands (split by the SO interaction) induced by external field might lead to a drastic change in the low energy properties of superconductivity. More specifically, considering the Dresselhaus-type interaction appropriate for the crystal symmetry of $I4_3d$, the energy gap along the Fermi momentum $k_F \parallel (001), (111), \text{and} (100)$, for example, is $\sqrt{(\mu_B H_z)^2 + \Delta_2^2} - \mu_B H_z$, so that it may be reduced to zero when $\mu_B H_z \gg \Delta$ [where $\Delta$ is the gap energy at zero field, $H_z$ is the external field applied along (001) axis].[4] This means the occurrence of field-induced point nodes that would lead to enhanced quasiparticle excitation. It must be noted, however, that the presence of double-gap in $Ln_2C_3$ may require a more careful examination for this effect, since the suppression of smaller gap (having a smaller upper critical field $H_{c2}$) by external field would show up in a similar way. Here, we report on the result of field-induced effect in La$_2$C$_3$ studied by muon spin rotation ($\mu$SR) experiment.

A conventional $\mu$SR experiment was carried out for a La$_2$C$_3$ sample ($T_c = 10.9$ K) on the M15 beamline of TRIUMF, Canada, where details on the experiment are described in the previous report.[2] The sample was common to the previous experiment, which turned out to be a typical double-gap superconductor [$\Delta_1(0) = 2.7(1)$ meV, $\Delta_2(0) = 0.6(1)$ meV]. For the field-scan measurements at 2 K (where both gaps are present), the sample was cooled down to the target temperature after the external magnetic field was stabilized at a temperature above $T_c$ in order to minimize the effect of flux pinning. Fig. 1 shows some examples of $\mu$SR time spectra obtained under a field of 0.25 and 4.0 T, where open symbols show the data in the normal state. While the enhancement of spin relaxation upon cooling down to 2 K is observed for both cases (filled symbols), it is also noticeable that the relaxation in the normal state is enhanced by increasing external field. Considering that La$_2$C$_3$ has a cubic structure (bcc, $I4_3d$) and that it is free from any local d or f electrons, we can attribute this field-induced magnetization uniquely to the van Vleck-like paramagnetism due to the SO interaction.

The spin relaxation rate in the normal state is deduced from fits using a Gaussian damping

$$A(t) = A_0 \exp(-\sigma_n^2 t^2) \cos(\gamma \mu_B t + \phi), \quad (1)$$

where $A_0$ is the initial asymmetry, $\gamma \mu_B$ is the muon gyromagnetic ratio ($=135.53$ MHz/T), $B$ is the local field felt by muons ($\approx \mu_0 H$, with $H$ being the external field), and $\phi$ is the initial phase of rotation. Although the fit with Eq. 1 does not reproduce data at higher fields (particularly above 3 T) where the spectra exhibit a tendency toward an exponential damping, we resort to this simple form for the convenience of evaluating qualitative trend for $\sigma_n$ versus
field. As shown in Fig. 2, $\sigma_n$ exhibits a quasi-linear dependence on the external field. This is understood by considering the field-induced van Vleck-like susceptibility ($\chi^v$) whose magnitude depends on the direction of the primary axis for the SO interaction, so that it may lead to an inhomogeneity of effective field in polycrystalline sample,

$$\sigma^2_n = \gamma^2 \mu H^2 (\chi^v)^2 + \sigma^2_v,$$

(2)

where $\sigma_0$ is the contribution of random local fields from $^{139}$La nuclear magnetic moments. (More specifically, $\chi^v$ also includes the Pauli paramagnetic term from the SO subbands.) The slight change of $\sigma_n$ observed around 3–4 T might be an artifact due to the deteriorated quality of fit with Gaussian damping. We also note that $\sigma_n$ is mostly independent of temperature between 11 K and $\sim$ 150 K under a field of 0.25 T.

Considering the contribution of field inhomogeneity due to the van Vleck-like paramagnetism, we deduce the spin relaxation rate in the mixed state by fits of $\mu$SR time spectra using a form,

$$A(t) = A_0 G^n(t; B) \exp \left[ -\frac{1}{2} \sigma^2_v t^2 \right] \cos(\gamma_0 B t + \phi),$$

(3)

where, instead of eq. (1), $G^n(t; B)$ is chosen to best reproduce the spectra at 15 K (> $T_c$) at each field. [$G^n(t; B)$ consists of a sum of two exponential damping signals, where the parameters describing $G^n(t; B)$ is determined by the spectra at 15 K at each field and then fixed to these values for the fit of spectra at 2 K to extract $\sigma_v$ reliably.] In this definition, $\sigma_v$ corresponds to the second moment for the field distribution $[B(r)]$ in the mixed state,

$$\sigma^2_v = \gamma^2 (\langle B(r) - \mu_0 H \rangle)^2.$$

(4)

Fig. 3 shows the deduced value of $\sigma_v$ at respective fields, where one can clearly observe a trend of steeper reduction with increasing field at lower fields (below $\sim$ 3 T).

In the limit of extreme type II superconductors [i.e., $\lambda/\xi \gg 1$, where $\lambda$ is the effective London penetration depth and $\xi = \sqrt{\Phi_0/(2\pi H_c2)}$ is the Ginzburg-Landau coherence length, $\Phi_0$ is the flux quantum, and $H_c2$ is the upper critical field], $\sigma_v$ is determined by $\lambda$ using a relation,[5]

$$\sigma_v(h) = 0.0274 \frac{\gamma^2 \Phi_0}{\lambda^2} (1 - h) \sqrt{1 + 3.9(1 - h)^2}$$

(5)

where $h$ is the field normalized by the upper critical field ($h = H/H_c2$). Although Eq. 5 exhibits a tendency of concave curve, it does not reproduce the field dependence of $\sigma_v$ observed in Fig. 3. This is particularly true when the known value of $H_{c2} (\sim 13$ T at 2 K) is considered; the situation is illustrated by a dashed curve in Fig. 3.

Since La$_2$C$_3$ is known to have a double-gap structure in the order parameter, it is natural to expand Eq. 5 into the following form,

$$\sigma_v(h) = w\sigma_v(h_1) + (1 - w)\sigma_v(h_2),$$

(6)

where $h_i = H/H_{c2}^{(i)}$, and $H_{c2}^{(i)}$ is the upper critical field corresponding to the respective energy gap [$\sigma_v(h_i)$ must be set to zero for $h_i > 1$]. The linear combination of two components is appropriate, as $\sigma_v$ is proportional to the superfluid density (i.e., $\sigma_v \propto n_s$). The solid curve in Fig. 3 is the best fit with Eq. 6, which yields $\lambda = 390(3)$ nm, $H_{c2}^{(1)} = 16(4)$ T, $H_{c2}^{(2)} = 3.6(4)$ T, and $w = 0.41(9)$.

It is interesting to note that the ratio of the upper critical field [$H_{c2}^{(1)}/H_{c2}^{(2)} = 4.4 \pm 1.4$] is comparable with that of the two energy gaps [$\Delta_1(0)/\Delta_2(0) = 4.5(1)$], and that the relative weight $w$ also agrees with that deduced from the temperature dependence of $\sigma_v$ [where $w = 0.38(2)$].[2] Provided that the physical parameters obtained at 2 K is not far from those at $T = 0$, it might be allowed to discuss the relation between the BCS coherence length and energy gap based on the equation,

$$\xi_0 = \frac{\hbar v_F}{\pi \Delta(0)} \simeq \xi, \ (T \rightarrow 0)$$

(7)

where $v_F$ is the Fermi velocity. Since the upper critical field is proportional to the inverse squared of $\xi$ [$H_{c2} = \Phi_0/(2\pi \xi^2)$], one may expect $H_{c2} \propto \Delta^2(0)/v_F^2$. Thus, the observed coincidence between the ratios of $H_{c2}$ and of $\Delta(0)$ may imply that the Fermi velocity also varies between bands nearly by a factor of two.

Another possible reason for the steeper reduction of superfluid density is the field-induced nodes in the energy gap and associated quasiparticle excitation (QP) specific to noncentrosymmetric superconductors.[8] The characteristic field, $H_{so}$, may be provided in relation to the gap,

$$H_{so}^{(i)} \sim \frac{\Delta_i(0)}{\mu_B},$$

(8)

which yields $H_{so}^{(1)} = 47(2)$ T and $H_{so}^{(2)} = 10(2)$ T. It is predicted in the calculation of electronic specific heat that the QP excitation is strongly enhanced to
reduce the superfluid density towards a field 0.55-0.7H_{so} (depending on the magnitude of the coherence length). Both of two corresponding fields, however, seem to be too high to explain the characteristic field of kink observed in Fig. 3 (≃ 3 T). On the other hand, it is also predicted for the case of small SO interaction (compared with Δ(0)) that the QP excitation might be enhanced below ∼ 0.1H_{so}, primarily due to the asymmetric deformation of the Fermi surface. In this case, both of the characteristic fields (≃ 4.7 T and 1.0 T) are not far from that observed in Fig. 3 and the weighted average (≃ 2.5 T) turns out to be in good accord with the kink of \( \sigma_v \).

It is presumable as an actual situation that the field dependence of \( \sigma_v \) reflects the effect of double-gap as well as that of the SO interaction, and they are not discernible within the present resolution of data along the magnetic field.

In conclusion, we demonstrated the presence of spin-orbit interaction by showing the occurrence of field-induced enhancement of muon spin relaxation in the normal state of a noncentrosymmetric superconductor, La$_2$C$_3$. In the mixed state of La$_2$C$_3$, we also showed that the field dependence has a clear kink around 3 T, and that this feature may be well explained by considering the effect of i) double-gap structure in the order parameter previously established by the temperature dependence of \( \sigma_v \), and ii) that of the spin-orbit interaction leading to the field-induced deformation of the Fermi surface.

We would like to thank the staff of TRIUMF for their technical support during the \( \mu \)SR experiment.

We also thank S. Fujimoto for helpful discussion on the effect of noncentrosymmetry on the electronic property of sesquicarbides. This work was supported by the KEK-MSL Inter-University Program for Oversea Muon Facilities and a Grant-in-Aid for Scientific Research on Priority Areas by Ministry of Education, Culture, Sports, Science and Technology, Japan.

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