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Superconductivity in \( Y_2C_3 \) with medium \( T_c \)

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Abstract

Recently, we have reported a relatively high-\( T_c \) superconductivity in \( Y_2C_3 \) at 18 K whose \( T_c \) could be changed by synthesis conditions from 10 to 18 K [G. Amano, S. Akutagawa, T. Muranaka, Y. Zenitani, J. Akimitsu, Superconductivity at 18 K in yttrium sesquicarbide system, \( Y_2C_3 \) \( J. \) Phys. Soc. Jpn. 73 (2004) 530.] We synthesized a high-purity sample of the medium-\( T_c \) phase in \( Y_2C_3 \) and examined its physical properties in detail. From a specific heat measurement, \( \Delta C(T)/\gamma T_c \) value is calculated to be 6.50 and the superconducting gap is estimated to be \( 2\Delta/k_B \) \( T_c = 5.40 \), indicating that the superconductivity in \( Y_2C_3 \) can be described by an \( s \)-wave strong coupling regime.

Keywords: Superconductivity; Carbide; \( Y_2C_3 \); Magnetic susceptibility; Specific heat

1. Introduction

The rare earth carbide system has been attracting renewed interests since the discovery of superconductivity in the layered yttrium carbide halides \( Y_2C_2I_2 \) (\( T_c = 9.97 \) K) and \( Y_2C_2Br_2 \) (\( T_c = 5.04 \) K) [1]. Binary and quasi-binary yttrium carbides (in particular Th-doped yttrium carbide (\( Y_{1-x}Th_x \)) \( C_2 \) \( T_c = 17.0 \) K) [2] have also attracted considerable attention since their \( T_c \) values are close to those of A15 compounds [3,4]. Recently, our group has reported the discovery of superconductivity at 18 K in \( Y_2C_3 \) [5]. In a previous study, Novokshonov showed that \( Y_2C_3 \) crystallizes in a body-centered cubic (BCC) \( Pm\overline{3}m \) type structure and \( I4d \) space group with eight formula units per unit cell by X-ray diffraction investigation [6].

The crystal structure of \( Y_2C_3 \) is shown in Fig. 1. In this structure, \( Y \) atoms are aligned along the \( \{111\} \) direction and \( C \) atoms form dimers. The superconductivity of yttrium sesquicarbide was found by the Krupka group [7]. They claimed that phase composition has a significant effect on \( T_c \) over the entire homogeneity range. The magnetic susceptibility indicated that the material was superconducting over the entire homogeneity range with \( T_c \) ranging from 6 to 11.5 K. To increase \( T_c \), we synthesized this material under high temperature and high pressure conditions, and consequently succeeded in synthesizing the high-\( T_c \) phase of \( Y_2C_3 \). From a theoretical point of view, Shein et al. reported the band structure of \( Y_2C_3 \) [8]. The band structure shows that the hybridization of \( C-C \) dimer antibonding and the \( Y-4d \) characteristics are dominant at the Fermi level. The electronic structure of other metals with \( C \) dimers both at the tight binding [9] and density functional levels [10] suggests that the electronic structure could have substantial \( C-C \) antibonding character near the Fermi level. Moreover, Singh et al., reported the magnitude of the electron-phonon coupling in \( Y_2C_3 \) from density functional calculations of the electronic structure using the general potential linearized augmented plane wave (LAPW) method in their recent report [11]. They suggest that low-frequency metal atom vibrations have the largest electron-phonon coupling whereas the contribution of the high-frequency \( C-C \) stretching vibrations is found to be comparatively small.

It is interesting to have a deep insight into the mechanism of superconductivity in \( Y_2C_3 \). From an experimental point of view, Nakane et al., examined the reproducibility of superconductivity in \( Y_2C_3 \) and succeeded in transport measurements after our report [12]. Because the upper critical field \( (H_{c2}) \) of this material was estimated to be approximately 30 T from their report and its \( T_c \) relatively high among simple binary intermetallic compounds, \( Y_2C_3 \) has attracted considerable attention. However, there still remains much to be understood on the mechanism of superconductivity because of the difficulty to synthesize pure samples and to stabilize the \( Y_2C_3 \) phase in air. We established a technique for synthesizing a high-purity sample. In this paper, we report on the physical properties of the medium-\( T_c \) phase (\( -15 \) K) of \( Y_2C_3 \).
2. Experimental details

The samples were prepared by mixing appropriate amounts of Y (99.9%) and C (99.95%) powders in a dry box and synthesized by arc melting in high purity-Ar gas. Binary alloys were in the first stage prepared by arc melting, after which they were subjected to high-pressure and high-temperature treatment to produce the sesquicarbide phase. All subsequent sample manipulations, including preparations for high-pressure experiments, were performed in a dry He atmosphere. The melted samples were heated to 1400–1600 °C for 5 min and maintained for 10–30 min under a pressure of 4–5.5 GPa using a cubic-anvil-type high-pressure apparatus and quenched to room temperature within a few seconds. The samples were examined by powder X-ray diffraction analysis, using a conventional X-ray spectrometer with a graphite monochromator (RINT-1100 RIGAKU). Intensity data were collected with CuKα radiation over a 2θ range from 5 to 80° at 0.02° step widths. The magnetic susceptibility and magnetization measurements were performed with a SQUID magnetometer (MPMSR2 Quantum Design Co., Ltd) and a PPMS system (Quantum Design Co., Ltd). The specific heat measurement was performed using the PPMS system (Quantum Design Co., Ltd) in the temperature range of 2–30 K.

3. Experimental results

Fig. 2 shows the powder X-ray diffraction pattern at room temperature of Y2C3. The Y2C3 phase was obtained as the main phase and indexed as a cubic unit cell with the space group I43d. The lattice constant calculated from all indices is 8.198 Å. Weak additional reflections were observed, which are attributed to an impurity phase of Y2OxCy, whose overall fraction is less than 6%.

The superconductivity of Y2C3 was confirmed by DC susceptibility in a magnetic field of 10 Oe, as shown in Fig. 3. The magnetic susceptibility of Y2C3 significantly decreases at 14.6 K, suggesting the occurrence of superconductivity. The volume fraction of superconductivity at 1.8 K is estimated to be approximately 27% in the field cooling process. Fig. 4 shows magnetization vs. magnetic field (M–H) curves, exhibiting the behavior of a typical type-II superconductor. From the M–H curves, we estimated the lower critical field, $H_{cl}(T)$ defined as a magnetic field where the initial slope meets the extrapolation curve of $(M_{up} + M_{down})/2$. The roughly estimated $H_{cl}(0)$ of Y2C3 is 3.5 mT. The penetration depth $\lambda(0)$ is calculated to be approximately 4300 Å from the relationship between $H_{cl}(0)$ and $\lambda(0)$, $\mu_0H_{cl} = \phi_0/\pi\lambda^2$, where $\mu_0$ and $\phi_0$ are the magnetic permeability in vacuum and quantum flux, respectively.

Fig. 5 shows the specific heat of Y2C3 taken at zero field and 8 T. As shown in Fig. 5, the specific heat taken at zero field
shows the \( Q \) and \( D \), respectively. The Debye temperature \( T_d \) determined from the susceptibility data.

Fig. 4. Magnetization vs. magnetic field \((M-H)\) curves of \( Y_2C_3 \). The inset shows the \( M-H \) curves in the low-field region. exhibits a sharp jump of \( \Delta C (T_c)=183 \text{ mJ/mol K} \) at \( T_c=14.6 \text{ K} \), with a width of 0.6 \( \text{ K} \), exactly the same as the \( T_c \) determined from the susceptibility data. \( T_c \) is defined as the midpoint of the jump. We assume that the overall specific heat is composed of the electron and lattice parts, \( C(T)=C_{el}(T)+C_{ph}(T) \). The lattice part is expressed by the \( \beta T^3 + \delta T^4 \) term at low temperature below the Debye temperature \( \Theta_D \), and the electron specific heat \( C_{el} \) is expressed by the \( \gamma T \) term. Thus, the specific heat in the normal state can be expressed by the formula \( C(T)/T=\gamma + \beta T^2 + \delta T^4 \).

From the \( C(T)/T-1/T^2 \) plot in Fig.5, the \( \gamma \), \( \beta \) and \( \delta \) values of \( Y_2C_3 \) was obtained to be 3.03 \( \text{ mJ/mol K}^2 \), 0.20 \( \text{ mJ/mol K}^4 \) and \( 1.65 \times 10^{-4} \text{ mJ/mol K}^6 \) and, respectively. The Debye temperature \( \Theta_D \) is related to the coefficient of the \( T^3 \) term, which originates from the phonon contribution \( \beta = N(12/5)\pi^2 R\Theta_D^3 \), where \( R=8.314 \text{ (J/mol K)} \) and \( N=40 \). The \( \Theta_D \) of \( Y_2C_3 \) was calculated to be 728 K. \( \gamma \) and \( \Theta_D \) are larger than those of the previously reported low-\( T_c \) phase in \( Y_2C_3 \) \( (T_c=10.0 \text{ K}) \) [13]. Cort et al., reported that \( \gamma \) and \( \Theta_D \) were 2.8 \text{ mJ/mol K}^2 and 557 K, respectively. Fig. 6 shows the temperature dependence of \( C_{el} \) in \( Y_2C_3 \).

Fig. 5. Temperature dependence of specific heats in \( Y_2C_3 \). The dashed lines are fitting results.

known that \( \Delta C(T_c)/\gamma T_c \) obtained from a heat capacity measurement shows the strength of electron–phonon coupling and its value is 1.43 in the BCS weak-coupling limit. We obtained a \( \Delta C(T_c)/\gamma T_c \) of 6.50. Moreover, the fitting below \( T_c \) gives \( \exp(-1/T) \) dependence as predicted within the BCS theory rather than \( 1/T \) dependence as in superconductors with an anisotropic symmetry of superconductivity. These results indicate that the symmetry of the superconductivity of \( Y_2C_3 \) is that of an isotropic s-wave. From the fitting below \( T_c \), we estimated \( 2\Delta/k_BT_c \) to be 5.40, which means that the \( 2\Delta/k_BT_c \) of \( Y_2C_3 \) is larger than that of \( \text{MgB}_2 \) \( (2\Delta/k_BT_c = 3.8–4.3) \) [14]. From the \( \Delta C(T_c)/\gamma T_c \) and \( 2\Delta/k_BT_c \) values determined from specific heat measurement, the superconductivity in \( Y_2C_3 \) indicates a strong coupling regime.

Fig. 6. Temperature dependence of \( C_{el} \) in \( Y_2C_3 \). The dashed lines are fitting results.

4. Summary

We succeeded in synthesizing a high-quality sample of medium-\( T_c \) phase in \( Y_2C_3 \) \( (T_c=14.6 \text{ K}) \), and examined the physical properties in detail by magnetic susceptibility, magnetization and specific heat measurements. The magnetization \((M-H)\) curves show typical type-II superconducting behavior, and \( H_{c2}(0) \) and \( \lambda(0) \) are estimated to be 3.5 mT and 4300 Å, respectively. From specific heat measurement, \( \gamma \), \( \Delta C(T_c)/\gamma T_c \), \( \Theta_D \) and \( 2\Delta/k_BT_c \) are determined to be 3.03 \text{ mJ/mol K}^2, 6.50, 728 K and 5.40, respectively. We conclude from these results that the symmetry of superconductivity in \( Y_2C_3 \) can be described as that of an isotropic s-wave strong coupling regime.

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