Anisotropic superconductivity in layered silicides

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Abstract

CaAlSi is a layered silicide with the AlB2-structure and shows superconductivity below 7.7 K. The angular dependence of the upper critical field, \(H_{c2}(\theta)\), in CaAlSi shows a cusp-like anomaly, suggesting the presence of decoupled two-dimensional superconducting layers. Concomitant with this feature, a strong lattice modulation along the \(c\)-axis exists in this material. On the other hand, \(H_{c2}(\theta)\) in CaGaSi is well fitted by the anisotropic Ginzburg–Landau (GL) model, and the superstructure along the \(c\)-axis is absent. In order to get some insight into the origin of the anomalous \(H_{c2}(\theta)\) in CaAlSi, we have performed magneto-optical observations of the vortex penetration for fields applied parallel and perpendicular to the \(c\)-axis. Vortices are found to penetrate the crystal inhomogeneously when the field is applied perpendicular to the \(c\)-axis, indicating the presence of macroscopic inhomogeneities in CaAlSi. We have also grown single crystals of CaAl\(_{1-x}\)Ga\(_x\)Si and studied the evolution of anomalous \(H_{c2}(\theta)\) and the lattice modulation along the \(c\)-axis. With increasing Ga content in CaAl\(_{1-x}\)Ga\(_x\)Si, the superstructure along the \(c\)-axis survives up to \(x = 0.2\), although the anomaly in \(H_{c2}(\theta)\) is strongly suppressed. It may suggest irrelevance of the two features, although it seems to be closely correlated in CaAlSi and CaGaSi. Possible scenarios for the appearance of the anomalous \(H_{c2}(\theta)\) are proposed.

Keywords: CaAlSi; Silicide; Superstructure

1. Introduction

The discovery of superconductivity in MgB\(_2\) with \(T_c \approx 39\) K [1] triggered the revival of research on intermetallic superconductors, in particular, the anisotropic compounds. Several new layered intermetallic superconductors have been reported since then [2–5]. AMSi family of pseudo-ternary layered silicides is a typical example, where \(A = \text{Ca, Sr or Ba} \) and \(M = \text{Al or Ga} \). We have successfully grown single crystals of CaAlSi and reported fundamental physical properties of this materials [6]. One of the most remarkable features of CaAlSi is the anomalous angular dependence of the upper critical field, \(H_{c2}\). The presence of a cusp-like feature when the field is applied parallel to the superconducting plane suggests the presence of decoupled two-dimensional superconducting layers in this material [7]. At the same time, CaAlSi has a superstructure along the stacking direction of the \(c\)-axis.

Similar studies on CaGaSi single crystals show interesting contrast. In CaGaSi, the angular dependence of \(H_{c2}\) is well described by the anisotropic Ginzburg–Landau (GL) model and there is no superstructure in this material [8]. In order to investigate the evolution of the anomalous angular dependence of \(H_{c2}\) and the superstructure from CaAlSi and CaGaSi, we have grown single crystals of CaAl\(_{1-x}\)Ga\(_x\)Si and studied their anisotropic physical properties and structure.

Even if we consider the superstructure of CaAlSi along the \(c\)-axis and the presence of the normal layer, its thickness can never be large enough to decouple superconductivity. This means that we need to consider inhomogeneity in much larger scale. Possible inhomogeneities beyond 1 \(\mu\)m scale can be visualized using magneto-optical (MO) technique. We have observed the vortex penetration into the \(ab\)-plane and the \(ac\)-plane of CaAlSi crystal and found an inhomogeneous penetration along the superconducting plane, which may be responsible for the decoupling of the two-dimensional superconducting layers.
2. Experiments

Single crystals of CaAl$_{1-x}$Ga$_x$Si (x = 0, 0.1, 0.2, 0.5, 1) are grown by the floating-zone method using an image furnace as described in Ref. [6]. After the growth, crystals are annealed in an evacuated fused-quartz tube with a chunk of calcium at 700 °C for 24 h to remove the residual strain and make the superconducting transition sharper. Electrical resistivity is measured by the conventional four-probe method using an AC resistance bridge (LR-700, Linear Research). The upper critical field is determined either by the onset of diamagnetism measured by a SQUID magnetometer (MPMS-XL5, Quantum Design) or by the resistive transition as a function of magnetic field at various temperatures and field-angles using a horizontal superconducting magnet. The superstructure is studied using a four-circle X-ray diffractometer (AFC-7PC, Rigaku). MO observations are used to check the possible inhomogeneities of the crystal. Polished surfaces the $ab$-plane and the $ac$-plane of a CaAlSi single crystal (~900 × 900 × 500(μm)$^3$) are examined by putting an in-plane magnetized garnet film as a magnetic-field indicator [9]. The light intensity modulated by the local magnetic induction on the sample surface is visualized using a cooled CCD camera (6E, Apogee).

3. Results and discussion

Fig. 1(a) shows the temperature dependence of the resistivity in CaAlSi for currents passing along the $ab$-($\rho_{ab}$) and the $c$-axis ($\rho_c$). Both $\rho_{ab}$ and $\rho_c$ are measured on the same piece of the crystal by re-defining the contact pads for each measurement. $\rho_{ab}$ shows a linear temperature dependence in a wide temperature range with residual resistivity of about 10 μΩ cm and residual resistivity ratio of about 3. The anisotropy of the resistivity $\rho_c/\rho_{ab}$ is about 5 in this crystal and typically ranges between 3 and 5. $\rho_c$ shows a tendency of saturation near room temperature, as in the case of metals with relatively large resistivity. Fig. 1(b) shows a blow-up of the low temperature part of Fig. 1(a) showing the superconducting transition. Both $\rho_{ab}$ and $\rho_c$ shows a clear transition to a superconducting state. However, it should be noted that the apparent transition temperature for $\rho_c$ is slightly lower than $\rho_{ab}$. This fact suggests that the superconducting transition temperature has a distribution along the $c$-axis.

Fig. 2(a) shows the temperature dependence of $H_c2$ in CaAlSi determined by the onset of diamagnetism measured by the SQUID magnetometer for two directions. The temperature dependence of $H_c2$ near $T_c$ is linear for both directions. The anisotropy of $H_c2$, $H_{c2}^{ab}/H_{c2}^{ac}$, is about 2, consistent with the anisotropy of the resistivity, $H_{c2}^{ab}/H_{c2}^{ac} = \rho_c/\rho_{ab})^{1/2}$. $H_c2(\theta)$ is determined by measuring the resistive transition as a function of magnetic field at various field-angles and temperatures. Thus determined $H_c2(\theta)$ is shown in Fig. 2(b). Although the anisotropy is small, the $H_c2(\theta)$ cannot be described by the anisotropic GL model shown by the dotted lines. Instead, it is well fitted by the Tinkham model developed for a thin-film superconductor shown by the solid lines, although the sample is a bulk superconductor. The inset shows the $H_c2(\theta)$ determined by different criteria $\rho = 0.9 \rho_n$ (pluses), 0.5 $\rho_n$ (circles), 0.1 $\rho_n$ (crosses). It is clear that the cusp-like anomaly of $H_c2(\theta)$ is independent of the criterion to determine $H_c2$. We have also confirmed that the same anomalies appear when we determine $H_c2(\theta)$ using magnetic permeability [10]. To explain the anomalous $H_c2(\theta)$ the presence of the non-superconducting layer to decouple superconductivity is required. Detailed structural analyses of CaAlSi reveal the presence of superstructure along the $c$-axis with a period of either 5c or 6c [6]. Although the superstructure may allow the presence of normal layers, the possible maximum thickness of the normal layer 6c (~2.4 nm) is not thick enough to decouple two-dimensional superconductivity. This means that we need much larger scaled inhomogeneity in the crystal to explain the anomalous $H_c2(\theta)$.

Fig. 3(a) shows the $H_c2(\theta)$ in CaAl$_{1-x}$Ga$_x$Si (x = 0.2). Unlike CaAlSi, the $H_c2(\theta)$ does not show cusp-like anomaly. However, the X-ray diffraction intensity in the (1, 0, $\bar{t}$)-zone clearly shows the presence of superstructure along the $c$-axis. These two results strongly suggest that the
anomalous \(H_{c2}(\theta)\) and the superstructure along the \(c\)-axis is not directly related, although their relevance are implied when we compare CaAlSi and CaGaSi.

In order to clarify the possible presence of inhomogeneity in CaAlSi, we have made MO imagings of vortex penetration for two directions. Figs. 4(a)–(f) show MO images for the vortex penetration in CaAlSi when the field is applied along the \(c\)-axis. Vortices start to penetrate the crystal at \(H_a/2\text{Oe} (\text{Fig. 4(b)})\). As the applied field is increased further, penetrated vortices form nearly symmetric pattern (Figs. 4(c)–(e)). Finally, vortices penetrate the whole sample, forming double-Y shaped pattern. These symmetric patterns of penetrated vortices indicate the homogeneity of the sample for fields along the \(c\)-axis. Vortices start to penetrate the crystal at \(H_a/35\text{Oe} (\text{Fig. 5(b)})\). As the applied field is increased further, vortices penetrate the sample inhomogeneously (Figs. 5(c)–(f)). This suggests that the lower critical field has a large distribution along the \(c\)-axis, indicating the macroscopic inhomogeneity of the sample when the field is applied parallel to the \(ab\)-plane. The region with the lower critical field may correspond to the normal region, which decouple the superconducting region, giving rise to the cusp-like feature of \(H_{c2}(\theta)\).

Finally, let us discuss possible origins of the anomalous \(H_{c2}(\theta)\). The presence of the macroscopic inhomogeneity as revealed by the MO imaging supports the decoupled two-dimensional superconducting layers as the origin of anomalous \(H_{c2}(\theta)\). However, in such a case, the temperature dependence of \(H_{c2}\) is expected to be \(H_{c2}(T) \propto (1-T/T_c)^{1/2}\), inconsistent with the data shown in Fig. 2(a). One the other
hand, the critical field related to the surface conductivity \( H_{c3} \) can have a cusp-like anomaly for fields parallel to the superconducting plane. However, in such a case, the volume fraction of the superconductivity at the critical field is expected to be negligible, inconsistent with the bulk superconducting transition confirmed by the specific heat measurement for \( H_{Jab} \)-plane \[6\].

4. Summary

In CaAlSi, we find an anomalous angular dependence of the upper critical field \( H_{c2}(\theta) \) and a strong lattice modulation along the \( c \)-axis. On the other hand, \( H_{c2}(\theta) \) in CaGaSi is well fitted by the anisotropic GL model, and the superstructure along the \( c \)-axis is absent. However, in CaAl\(_{1-x}\)Ga\(_x\)Si \((x = 0.2)\), only the superstructure exists without anomalous \( H_{c2}(\theta) \), suggesting the irrelevance of these two features. We find a macroscopic inhomogeneity along the \( c \)-axis in CaAlSi by the MO imaging, which may be related to the presence of anomalous \( H_{c2}(\theta) \). We have proposed two scenarios for the anomalous \( H_{c2}(\theta) \), based on the decoupled two-dimensional superconducting layers or on the surface superconductivity. Although both scenarios can explain the anomalous \( H_{c2}(\theta) \), they still have a certain inconsistency with the experimental results.

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