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Superconductivity in the Ni-based ternary compound $\text{La}_6\text{Ni}_2\text{Sn}$

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Abstract

We have investigated the transport and magnetic properties of the ternary compound $\text{La}_6\text{Ni}_2\text{Sn}$ by means of the ac electrical resistivity ρ , dc magnetic susceptibility χ_{dc} , and specific heat $C(T)$ measurements. Bulk superconductivity was observed in this compound with a superconducting transition temperature of ~ 2.25 K as inferred from a sudden drop to zero value in the $\rho(T)$ curve, the diamagnetic behavior in the $\chi_{\text{dc}}(T)$ curve, and a jump in the $C(T)$ curve. Type II superconductivity can also be inferred from the isothermal M – H curve measured at $T = 2$ K. The initial slope of the upper critical magnetic field $-(dH_{c2}/dT)_{T_{c0}}$ has a value of 4000 Oe/K, from which a zero-temperature superconducting coherence length $\xi_0 = 230$ Å can be obtained. Analysis using the specific heat data yields a Debye temperature $\Theta_D = 135$ K for this compound.

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1. Introduction

The discovery of superconductivity in the R–Ni–B–C (R = Y or rare earth) systems has renewed a lot of research interest in the Ni-based compounds [1–3]. Studies of the rare-earth Ni-based ternary compounds are of special interest because superconductivity and various types of magnetic order have been observed in these systems [4–7]. The $\text{R}_6\text{Ni}_2\text{Sn}$ (R = rare earth ions) compounds are of the $\text{Ho}_6\text{Ni}_2\text{Ga}$ -type structure with space group Immm [8]. Among the $\text{R}_6\text{Ni}_2\text{Sn}$ compounds, $\text{Er}_6\text{Ni}_2\text{Sn}$ has been mostly studied

because of its usage for the magnetic refrigeration [9,10]. In this paper, we report the first observation of bulk superconductivity in the Ni-based ternary compound $\text{La}_6\text{Ni}_2\text{Sn}$ with a transition temperature of 2.25 K. In addition, isothermal magnetization measurements at $T = 2$ K reveal type II superconductivity in this compound.

2. Experimental

The polycrystalline samples of $\text{La}_6\text{Ni}_2\text{Sn}$ were prepared by arc-melting stoichiometric amounts of the constituent elements (La: 99.99%, Ni: 99.99%, Sn: 99.9999%) together on a water-cooled copper hearth in a Zr-gettered argon atmosphere. A two-stage procedure ($\text{La}_6 + \text{Ni}_2\text{Sn}$) was utilized with negligible weight loss ($< 0.5\%$). To improve the

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sample homogeneity, the as melted samples were subsequently wrapped in Ta foil, sealed in quartz tube in argon atmosphere, and annealed at 550 °C for 3 days. The ac electrical resistivity $\rho(T)$ measurements on bar-shaped samples were performed in a He⁴ cryostat between 300 K and 2 K using a four-wire ac impedance bridge operating at 16 Hz with silicon diode as the temperature sensor. The dc magnetic susceptibility measurements were performed in a commercial superconducting quantum interference device (SQUID) magnetometer from 2 to 300 K in various applied magnetic fields. The specific heat measurements were performed in a commercial calorimeter in the temperature range between 1.8 and 15 K in different applied fields H .

3. Results and discussion

The ac electrical resistivity ρ versus temperature T curve in zero magnetic field for La₆Ni₂Sn is plotted in Fig. 1(a) for $0 \leq T \leq 300$ K. Above the superconducting transition temperature, $\rho(T)$ curve exhibits typical characteristics of common metal and decreases monotonically with decreasing temperature with a RR ratio of 4.4 and a residue resistivity $\rho_r = 28.35 \mu\Omega \text{ cm}$. The relatively high value of the residue resistivity ρ_r indicates that the sample is probably not very homogeneous. At ~ 2.8 K, the resistivity starts to drop to zero and the sample becomes superconducting with the midpoint transition temperature $T_c(50\%) = 2.4$ K and a transition width $\Delta T_c = 0.58$ K at zero magnetic field. Shown in Fig. 1(b) are selected curves for ac electrical resistivity ρ as a function of temperature T in different applied magnetic fields H below 2.0 kOe. With increasing applied magnetic field, the transition curves shift to lower temperature and the transition widths increase. Finally, when the field gets higher than 2.0 kOe, the superconducting transition is lower than 2.0 K.

The temperature dependences of the zero-field cooled (ZFC) and field-cooled (FC) magnetization curves measured in a field of 20 Oe between 2 and 4 K for the La₆Ni₂Sn sample are plotted in Fig. 2(a). The occurrence of diamagnetic transition in the ZFC curve near 2.4 K reveals the occurrence of superconductivity in this compound, in consistent

Fig. 1. (a) The ac electrical resistivity ρ versus temperature T curve in zero magnetic field for La₆Ni₂Sn for $0 \leq T \leq 300$ K. (b) $\rho(T)$ curves in different applied magnetic fields H below 2.0 kOe.

with the result as obtained from the $\rho(T)$ curve. The diamagnetism is much weaker in the FC curve. Plotted in the inset of Fig. 2(a) is the temperature dependence of the magnetization of La₆Ni₂Sn measured in an applied field of 5 kOe. The very weakly temperature dependent susceptibility reveals that La₆Ni₂Sn is a Pauli paramagnet and there is no magnetic moment associated with the Ni ions. Fig. 2(b) shows the isothermal magnetization curve for La₆Ni₂Sn that was measured on increasing field at $T = 2$ K. Which reveals type II superconductivity in this compound.

The specific heat as a function of temperature taken in zero magnetic field for La₆Ni₂Sn is plotted in Fig. 3(a) for $0 \leq T \leq 8$ K. The inset shows the details of the $C(T)$ curves in various applied magnetic fields (all data were taken with the

Fig. 2. (a) Temperature dependences of the ZFC and FC magnetization curves measured in a field of 20 Oe between 2 and 4 K for La₆Ni₂Sn. The inset is the magnetization data measured in an applied field of 5 kOe. (b) The isothermal magnetization curve for La₆Ni₂Sn measured on increasing field at $T = 2$ K.

magnetic field applied at the lowest temperature). Shown in Fig. 3(b) is the C/T versus T^2 plot for this sample. The zero temperature value of the electronic specific heat coefficient $\gamma = 6.30$ mJ/mol La–K² is obtained and the value of β as obtained from the slope at low temperatures is 1.19 mJ/mol La–K⁴. The latter value corresponds to the Debye temperature $\Theta_D = 135$ K for this compound. The observed value of $\Delta C/\gamma T_c$ at zero field is about 0.66, which is smaller than 1.43 as predicted from the BCS theory. Nevertheless, the discontinuity in the $C(T)$ curve is in consistent with the results of $\rho(T)$ and $\chi(T)$ measurements and indicates that La₆Ni₂Sn undergoes bulk superconducting transition with $T_c \sim 2.25$ K. Works for improv-

Fig. 3. (a) The specific heat as a function of temperature taken in zero magnetic field for La₆Ni₂Sn for $0 \leq T \leq 8$ K, with the inset showing the details of the $C(T)$ curves in various applied magnetic fields. (b) C/T versus T^2 plot for La₆Ni₂Sn.

ing the sample homogeneity are currently in progress.

The upper critical field $H_{c2}(T)$ for La₆Ni₂Sn cannot be obtained because it requires measurements far below 1.8 K (the lowest temperature we can achieve). However, one can estimate the orbital critical magnetic field at zero temperature from the weak-coupling formula [11] $H_{c2}^*(0) = -0.693 T_{c0} (dH_{c2}/dT)_{T_{c0}}$. The initial slope of the upper critical magnetic field $-(dH_{c2}/dT)_{T_{c0}}$ has a value of 4000 Oe/K (data not shown here), and the obtained value of $H_{c2}^*(0)$ is ~ 6240 Oe. From this, the coherence length at zero temperature for this compound can be estimated from the relation [12] $\xi_0^2 = \Phi_0 / 2\pi H_{c2}^*(0)$, where $\Phi_0 = ch/2e = 2.07 \times 10^{-7}$ Oe cm² is the flux quantum. The obtained value of ξ_0 is ~ 230 Å.

The value of the transport mean free path l_{tr} for $\text{La}_6\text{Ni}_2\text{Sn}$ can be estimated from the normal state resistivity at low temperatures by means of the formula [13] $l_{tr} = (1.27 \times 10^4)/\rho_0(Z/\Omega)^{2/3}$, where l_{tr} is in cm, ρ_0 is in $\Omega\text{ cm}$, and (Z/Ω) is the density of the conduction electrons. As a rough approximation, we assume the conduction electrons are contributed by the trivalent La atoms. And the value of $(Z/\Omega) \sim 8.42 \times 10^{22}\text{ cm}^{-3}$ is obtained. In addition, the extrapolated zero temperature resistivity ρ_0 for this sample can be replaced by the residue resistivity ρ_r , which has a value of $\rho_0 = 28.35\text{ }\mu\Omega\text{ cm}$. The value of $l_{tr} \sim 23\text{ }\text{\AA}$ is then obtained. This value is considerably smaller than that of ξ_0 , suggesting that $\text{La}_6\text{Ni}_2\text{Sn}$ is a dirty superconductor.

The conduction electron effective mass m_{eff} for $\text{La}_6\text{Ni}_2\text{Sn}$ can be calculated from the equation [14] $m_{\text{eff}} = \hbar^2 k_F^2 \gamma / 4\pi^4 (Z/\Omega) k_B^2$. Here γ is the electronic specific heat coefficient for $\text{La}_6\text{Ni}_2\text{Sn}$ which has a value of $6.30\text{ mJ/mol La-K}^2$ as obtained from the $C(T)$ measurement. The Fermi wave vector k_F can be estimated (with the assumption of a spherical Fermi surface in the sample) from the relation $k_F = (3\pi^2 Z/\Omega)^{1/3}$ and yields a value of $k_F = 1.36 \times 10^8\text{ cm}^{-1}$. From this, we then obtain $m_{\text{eff}} = 2.4m_e$, where m_e is the mass for the free electron.

In summary, we have performed the ac electrical resistivity ρ , dc magnetic susceptibility χ_{dc} , and specific heat $C(T)$ studies on $\text{La}_6\text{Ni}_2\text{Sn}$. We found that is a type II superconductor with $T_c = 2.25\text{ K}$ and $-(dH_{c2}/dT)_{T_c} = 4000\text{ Oe/K}$. The obtained value of transport mean free path l_{tr} is $23\text{ }\text{\AA}$, which is much smaller than the value of ξ_0 and implies dirty superconductivity in this compound. The Debye temperature for this compound is about 135 K and the effective mass of the conduction electrons is about $2.4m_e$ as estimated from the $C(T)$ data.

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