UPPER CRITICAL MAGNETIC FIELD AND SPECIFIC HEAT STUDIES OF LaCu

J. W. Chen

Department of Physics,
National Taiwan University, Taipei, Taiwan, R. O. C.

and

Y. Y. Chen

Institute of Physics, Academia Sinica, Taipei, Taiwan, R. O. C.

(Received 25 February 1994 by P. Wachter)

We have performed the upper critical magnetic field $H_{c2}(T)$ and specific heat $C(T)$ studies of LaCu. The initial slope of $H_{c2}(T)$ near the zero field superconducting transition temperature $T_c = 6.15$ K has a value of 2.97 kG/K, from which a zero-temperature superconducting coherence length $\xi_0 \approx 162$ Å can be inferred. Analysis using the specific heat data yields an effective mass of 4 m_e for the conduction electrons and a Debye temperature $\Theta_D = 148$ K for this compound.

I. Introduction

There have been a lot of research activities about the interaction between superconductivity and magnetism in the past few decades. Early experiments involved dissolving small amounts of magnetic ions into a superconducting matrix, such as $\text{La}_{1-x}\text{Gd}_x$ ^{1,2} and $(\text{La}_{1-x}\text{Gd}_x)\text{Al}_2$ ³, and demonstrated the destructive effect of the magnetic ions on the superconducting transition temperature T_c . Moreover, interesting phenomena between the magnetic ordering and superconductivity were observed in many systems. For example, in $(\text{Ce}_{1-x}\text{Ho}_x)\text{Ru}_2$ ⁴⁻⁶, superconductivity was observed to coexist with short range magnetic ordering at $x = 0.27$; reentrant superconductivity was observed in $(\text{La}_{1-x}\text{Gd}_x)\text{Ru}_2$ near $x = 0.05$ wherein the sample becomes superconducting at a temperature T_1 and returns to the normal state at a lower temperature T_2 at which short-range spin glass type of magnetic ordering forms in the sample⁷. Later discovery of the Chevrel phase compounds^{8,9} and the RRh_4B_4 (R = rare earth ions) compounds^{10,11} has renewed the interest in the question between superconductivity and long range magnetic ordering because the R ions in these systems occupy a periodic array in the lattice^{12,13}. Among the large varieties of R -Cu systems, RCu have been reported to crystallize in FeB -type structure for $R = \text{La}, \text{Ce}, \text{Pr}, \text{Nd}$, and Yb ¹⁴⁻¹⁶. An ac susceptibility study of LaCu indicates superconducting transition occurs at $T \approx 5.85$ K¹⁷. An antiferromagnetic transition was observed in CeCu with a Néel temperature T_N ranging from 2.7 K to 3.6 K^{15,18}. Magnetization measurements of PrCu and NdCu reveal anomalous characteristics in these compounds¹⁶. Recently we found that NdCu becomes antiferromagnetic with $T_N \approx 10$ K and a weak antiferromagnetic transition occurs in PrCu at $T = 13$ K¹⁹. Therefore it is interesting to investigate the effect of various types of magnetic ordering on the superconductivity in the $\text{La}_{1-x}\text{R}_x\text{Cu}$ ($R = \text{Ce}, \text{Pr}, \text{and Nd}$) systems. Before doing this, it is necessary to have an understanding of the normal state and superconducting state properties of the superconducting matrix LaCu. In this paper, we report the upper critical magnetic field $H_{c2}(T)$ and the specific heat $C(T)$ studies of the LaCu compound.

II. Experimental Details

The polycrystalline samples of LaCu were prepared by arc-melting the high purity elements (La: 99.99 %, Cu: 99.999 %) together on a water cooled copper hearth in a Zr gettered argon atmosphere, with no subsequent annealing. The $H_{c2}(T)$ data in this study were determined from the ac electrical resistivity $\rho(T)$ measurements on a bar-shaped sample performed in a helium dewar using a four-wire resistance bridge operating at a frequency of 16 Hz. A calibrated Ge thermometer was used for the zero field measurement and for the calibration of a carbon glass thermometer which was used as the thermometer for the non-zero field measurements up to $H = 15$ kG in this study. Dc magnetization measurement was performed using Quantum Design MPMS SQUID magnetometer from 2 K to 10 K in an applied magnetic field of 10 G. The specific heat $C(T)$ measurement was performed on a small LaCu sample in a He^3 - He^4 dilution refrigerator using the relaxation method in the temperature range of $0.3 \text{ K} \leq T \leq 16 \text{ K}$.

III. Results and Discussion

The ac electrical resistivity ρ versus temperature T curve in zero magnetic field for LaCu are plotted in Fig. 1(a) for $0 \text{ K} \leq T \leq 300 \text{ K}$. Above the superconducting transition temperature, $\rho(T)$ curve exhibits typical characteristics of common metals and can be expressed in terms of $\rho(T) = \rho_i + \rho_{ph}(T)$ with a residue resistivity $\rho_i = 1.33 \mu\Omega\text{-cm}$ and the phonon contribution to the resistivity $\rho_{ph}(T)$ can be fitted by the semi-empirical Bloch-Grüneisen formula using a Debye temperature $\Theta_D = 120$ K and a electron-phonon interaction constant C of $\approx 0.25 \mu\Omega\text{-cm/K}$. The large value of the RR ratio $(\rho(300 \text{ K})/\rho(7 \text{ K}) = 57)$ reflects high sample quality. At $T \approx 6.4$ K, the resistivity drops rapidly to zero and the sample becomes superconducting with the midpoint transition temperature T_c (50 %) = 6.15 K and a transition

Fig. 1(a). The ac electrical resistivity ρ as a function of temperature T in zero magnetic field for LaCu. 1(b). Selective curves for the ac electrical resistance R as a function of temperature T in different applied magnetic fields H below 15 kG.

width ΔT_c of 0.2 K. Dc magnetization measurement at $H = 10$ G reveals diamagnetic transition occurring at the same temperature and indicates bulk superconductivity in this sample. Shown in Fig. 1(b) are selected curves for ac electrical resistance R as a function of temperature T in different applied magnetic fields H below 15 kG. The normal state resistivity increases slightly with increasing applied magnetic field H which becomes more pronounced for $H \geq 6$ kG. Moreover, with increasing magnetic field, the superconducting transition curves shift to lower temperature and the transition widths increase. Finally, when the field gets higher than 12 kG, the superconducting transition temperature is lower than 2.5 K.

Shown in Fig. 2(a) are the heat capacity C divided by temperature T vs T data for LaCu. A jump in C is observed near $T = 5.8$ K, which is close to the value of T_c observed in the ac electrical resistivity measurement. The observed value of $\Delta C/\gamma T_c$ is about 0.25, which is substantially smaller than 1.43 for the weak coupling superconductors. Nevertheless, this jump in C indicates bulk superconductivity in the sample. The $C(T)/T$ vs T^2 data for LaCu are displayed in Fig. 2(b). A marked departure from the linear relation above T_c is observed which indicates the Debye temperature Θ_D for LaCu is quite low since the low temperature approximation for heat capacity $C = \gamma T + \beta T^3$ holds only for $T/\Theta_D \ll 1$. The zero temperature value of γ is ≈ 10 mJ/mole-K² and the value of β as obtained from the slope at low temperatures is 1.18 mJ/mole-K⁴. From the value of β , the Debye

Fig. 2(a). The heat capacity C divided by temperature T vs T for LaCu. 2(b). $C(T)/T$ vs T^2 data for LaCu.

temperature can be calculated from the relation $\Theta_D = (1.946 \times 10^6 N/\beta)^{1/3}$ with N being the number of atoms per formula unit ($N = 2$ here). The obtained value of Θ_D is 148 K, which is close to the value of Θ_D deduced from the $\rho(T)$ fitting.

The upper critical magnetic field as a function of temperature $H_{c2}(T)$ for LaCu determined from the ac electrical resistivity measurements are plotted in Fig. 3 with the solid circles represent T_c (50 %)'s, the horizontal bars represent the transition widths and the arrows at high fields indicate the transition is not complete at $T \approx 2.5$ K. The overall shape of the $H_{c2}(T)$ curve is typical of a conventional type II superconductor. The observed value of $H_{c2}(T = 0)$ is ≈ 12.6 kG, which is an order of magnitude lower than the obtained value of the paramagnetic critical field H_p ($H_p = 18.4 \times T_c(H = 0)$ kG ≈ 113 kG) and indicates that the orbital critical magnetic field $H_{c2}^*(T)$ is mainly responsible for the $H_{c2}(T)$ in this sample. The linear portion of the H_{c2} vs T curve at low magnetic fields has a slope $(-dH_{c2}/dT)_{Tc0} = 2.97$ kG/K which can be used to calculate the zero-temperature orbital critical magnetic field $H_{c2}^*(0)$ using the weak-coupling formula²⁰ $H_{c2}^*(0) = 0.693 \times (-dH_{c2}/dT)_{Tc} \times T_{c0}$ which yields a value of ≈ 12.6 kG. This value can be used to estimate the superconducting coherence length ξ_0 at $T = 0$ K through the relation²¹ $H_{c2}^*(0) = \Phi_0/2\pi\xi_0^2$, where $\Phi_0 = ch/2e = 2.07 \times 10^{-7}$ G-cm² is the flux quantum. The obtained value of ξ_0 is ≈ 162 Å.

Fig. 3. The upper critical magnetic field H_{c2} as a function of temperature T for LaCu. The solid circles represent data for T_c (50 %), the horizontal bars represent the transition widths, and the arrows at high fields indicate the transition is not complete at $T \approx 2.5$ K. The $H_{c2}(T)$ curve calculated from the WHHM theory are also plotted.

The value of the transport mean free path l_{tr} for LaCu can be estimated from its normal state resistivity at low temperatures. The extrapolated zero temperature resistivity ρ_0 for this sample is estimated to be $1.33 \mu\Omega\text{-cm}$. From this, l_{tr} can be calculated from the formula²² $l_{tr} = (1.27 \times 10^4)/\rho_0(Z/\Omega)^{1/3}$, 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 that the conduction electrons are contributed by the trivalent La atoms so that $Z = 12$ since there are four LaCu formula units per unit cell. From the lattice parameters for LaCu, we find $\Omega = 1.99 \times 10^{22} \text{ cm}^{-3}$. The value of $l_{tr} \approx 810 \text{ \AA}$ is then obtained. Since $\xi_0 \ll l_{tr}$, we suggest that LaCu is a clean superconductor.

The conduction electron effective mass m^* for LaCu can be estimated from the equation²³ $m^* = \hbar^2 k_F \gamma / 4\pi^4 (Z/\Omega) k_B^2$. Here γ is the electronic specific heat coefficient at low temperatures which has a value of 10 mJ/mole-K^2 as obtained from the $C(T)$ measurement and k_F is the Fermi wave vector which, with the assumption of a spherical Fermi surface in the sample, can be calculated from the relation $k_F = (3\pi^2 Z/\Omega)^{1/3}$ and yields a value of $1.21 \times 10^8 \text{ cm}^{-1}$. From this, we then obtain $m^* \approx 4 m_e$, where m_e is the free-electron mass.

The curve calculated from the Werthamer, Helfand, Hohenberg, and Maki (WHHM) theory^{24,25} of $H_{c2}(T)$ for the conventional type II superconductor using the experimental values of the initial slope $(-dH_{c2}/dT)_{Tc0}$ and T_{c0} for the minimum ($\lambda_{s0} = \infty$) paramagnetic limit, where λ_{s0} is the spin orbit scattering parameter, are also plotted in Fig. 3. The calculated $H_{c2}(T)$ curve fits the experimental data quite well in the whole temperature range. The calculated upper critical field at zero temperature $H_{c2}(0)$ is 22.6 kG .

In summary, we have performed the $H_{c2}(T)$ and $C(T)$ studies of LaCu. The initial slope $(-dH_{c2}/dT)_{Tc0}$ of this compound is of 2.97 kG/G . The calculated $H_{c2}(T)$ curve using the WHHM theory can fit the experimental data in the whole temperature range quite well. The obtained value of $l_{tr} \approx 162 \text{ \AA}$, which is much more than the value of ξ_0 and implies clean superconductivity in this system. The Debye temperature of this system is about 148 K and the effective mass of the conduction electrons is about $4 m_e$ as estimated from the $C(T)$ data. This work was supported by the R. O. C. National Science Council under Grant No. NSC 81-0208-M002-045.

References

1. B. T. Matthias, H. Suhl, and E. Corenzwit, *Phys. Rev. Lett.* **1**, 92 (1958).
2. R. A. Hein, R. L. Falge Jr., B. T. Matthias, and E. Corenzwit, *Phys. Rev. Lett.* **2**, 500 (1959).
3. M. B. Maple, *Physics Letters*, **26A**, 513 (1968).
4. B. T. Matthias, H. Suhl, and Corenzwit, *Phys. Rev. Lett.* **1**, 449 (1958).
5. W. Wilhelm and B. Hillenbrand, *J. Phys. Chem. Solids*, **31**, 559 (1970).
6. J. W. Lynn, D. E. Moncton, L. Passell, and W. Thomlinson, *Phys. Rev.* **B21**, 70 (1980).
7. T. F. Jones, J. F. Kwak, E. P. Chock, and P. M. Chaikin, *Solid State Commun.* **27**, 209 (1978).
8. R. Chevrel, M. Sergent, and J. Prigent, *J. Solid State Chem.* **3**, 515 (1971).
9. B. T. Matthias, M. Marezio, E. Corenzwit, A. S. Cooper, and H. Barz, *Science* **175**, 1465 (1972).
10. J. M. Vandenberg and B. T. Matthias, *Proc. Natl. Acad. Sci.* **74**, 1336 (1977).
11. B. T. Matthias, E. Corenzwit, J. M. Vandenberg, and H. Barz, *Proc. Natl. Acad. Sci.* **74**, 1334 (1977).
12. *Topics in Current Physics*, Vol. 32, eds. Ø. Fischer and M. B. Maple (Springer-Verlag, New York, 1982).
13. *Topics in Current Physics*, Vol. 34, eds. M. B. Maple and Ø. Fischer (Springer-Verlag, New York, 1982).
14. A. C. Larson and D. T. Cromer, *Acta Cryst.* **14**, 545 (1961).
15. R. E. Walline and W. E. Wallace, *J. Chem. Phys.* **42**, 604 (1965).

16. A. Iandelli and A. Palenzona, *J. Less-Common Metals*, 25, 333 (1971).
17. T. F. Smith and H. L. Luo, *J. Phys. Chem. Solids*, 28, 569 (1967).
18. E. Bauer, *Advance in Physics*, 40, 417 (1991).
19. J. W. Chen, *J. Magn. Mag. Mat.* in press.
20. R. R. Hake, *Appl. Phys. Lett.* 10, 186 (1967).
21. For example, see M. Tinkham, "Introduction to Superconductivity" (McGraw-Hill, New York, 1975).
22. T. P. Orlando, E. J. McNiff, S. Foner, and M. R. Beasley, *Phys. Rev.* B19, 4545 (1979).
23. For example, see C. Kittel, "Introduction to Solid State Physics", 6th edition, (John Wiley & Sons, New York, 1986).
24. N. R. Werthamer, E. Helfand, and P. C. Hohenberg, *Phys. Rev.* 147, 295 (1966).
25. K. Maki, *Phys. Rev.* 148, 392 (1966).