Superconductivity in the Ni-based ternary compound $LaNiGa_2$

N. L. Zeng and W. H. Lee*

Department of Physics, National Chung Cheng University, Ming-Hsiung, Chia-Yi 621, Taiwan, Republic of China

(Received 14 May 2002; revised manuscript received 30 July 2002; published 19 September 2002)

The agreement of the transition temperature $T_c = 1.93-2.01$ K (10–90 % values), measured by dc susceptibility and heat-capacity techniques, provides clear evidence of bulk superconductivity in $LaNiGa_2$. The normal-state specific-heat data can be fit to the expression $C_n = \gamma T + \beta T^3$ by a least squares analysis, where $\gamma = 11.64 \pm 0.05$ mJ/mol K^2 and $\beta = 0.332 \pm 0.06$ mJ/mol K^4, resulting in a Debye temperature $\Theta_D = 560 \pm 10$ K. Below T_c, the specific-heat data have a dominant low-temperature behavior of the form exp $[-\Delta(0)/k_B T]$, where the order parameter $2\Delta(0) = 3.2 k_B T_c$. In addition, the measured heat-capacity jump $\Delta C (= 32 \pm 2$ mJ/mol K) at the transition point is found to be equal to $1.40 \gamma T_c$, implying that $LaNiGa_2$ is a weakly coupled superconductor.

DOI: 10.1103/PhysRevB.66.092503 PACS number(s): 74.70.Ad, 74.25.−q

Superconductivity in nickel-based ternary or quartery intermetallic compounds was notably discussed recently.[1–9] As mentioned by Nagarajan *et al.*,[7] a relatively small number of nickel-containing superconductors is known in the literature. Clearly, the ferromagnetic element Ni in the compound usually has an adverse effect on the superconductivity. Thus the identification of superconducting Ni-based compounds is of high current interest. In this paper, we report the superconductivity in the compound $LaNiGa_2$. To our knowledge, this is the first nickel-based ternary-gallide superconductor. As shown by dc susceptibility and heat-capacity measurements, this compound undergoes a superconducting transition at 1.97 ± 0.02 K (the midpoint of the transition).

Polycrystalline $LaNiGa_2$ was synthesized by arc melting together with stoichiometric amounts of the constituent elements in a Zr-gettered arc furnace on a water-cooled Cu hearth under purified argon of about one atmosphere. La with a purity of >99.9% was obtained from the Materials Preparation Center of the Ames Laboratory. Ni with 99.9% purity and Ga with 99.99999% purity were purchased from Morton Thiokol, Inc. and Matthey Bishop, Inc., respectively. Due to sufficiently low vapor pressures of these elements at the melting temperature of the ternary compound, weight losses during several melting and turning cycles were less than 0.5%. The arc-melted sample was then wrapped in tantalum foil and zirconium foil, sealed under argon in a quartz tube, and annealed for ten days at 600 °C. This heat treatment was followed by a water quench to room temperature. A microcomputer controlled MXP3 diffractometer equipped with a copper target and a graphite monochrometor for Cu K_α radiation was used to obtain the powder x-ray diffraction (XRD) patterns. It was found that a nearly single phase sample could be achieved. As shown in Fig. 1, except for the two less intense reflections at $2\theta = 34.55°$ and 40.91°, each peak line of the XRD patern for $LaNiGa_2$ can be indexed in an orthorhombic $NdNiGa_2$-type structure[10] with space group C_{mmm}. Though the chemical formula of the impurity phase with the two unindexed peak lines in Fig. 1 is not clear, we still can identify the fact that the superconducting property of the sample investigated has no relation with this impurity phase which can also be found in our another nonsuperconducting La-Ni-Ga samples. A refinement of the lattice parameters of the unit cell was determined by the method of least squares using the eleven intense reflections for $2\theta < 50°$ and including an internal silicon standard ($a = 0.543083$ nm). The lattice parameters $\mathbf{a} = 4.268(1)$ Å, $\mathbf{b} = 17.70(1)$ Å, and $\mathbf{c} = 4.226(4)$ Å were then obtained, a little bit smaller than the previously reported values in the literature.[10]

Figure 2 displays the temperature dependence of the zero-field cooled and field-cooled magnetization for the $LaNiGa_2$ sample measured in a field of 10 Oe between 1.80 and 3.00 K with a Quantum Design superconducting quantum interference device magnetometer. It is seen that the 10–90 % values of the superconducting transition signal occur at the temperatures 1.93 and 2.01 K. The quite narrow transition width (0.08 K) is a manifestation of purity of the superconducting phase. The strong diamagnetic signal of χ_{dc} does not prove the existence of bulk superconductivity in $LaNiGa_2$. Convincing evidence for bulk superconductivity is a specific-heat anomaly.

The specific heat of a piece (~3 mg) cut from the sample was measured in the range of 0.35–20 K with a He^3 relaxation calorimeter using the heat pulse technique[11] in the

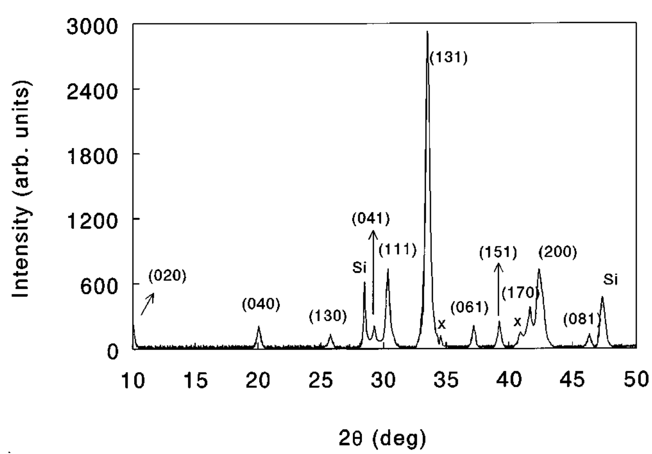

FIG. 1. Room-temperature powder-x-ray-diffraction pattern of $LaNiGa_2$ using Cu $K\alpha$ radiation. The two less intense peak lines with *x* mark belong to impurity phase.

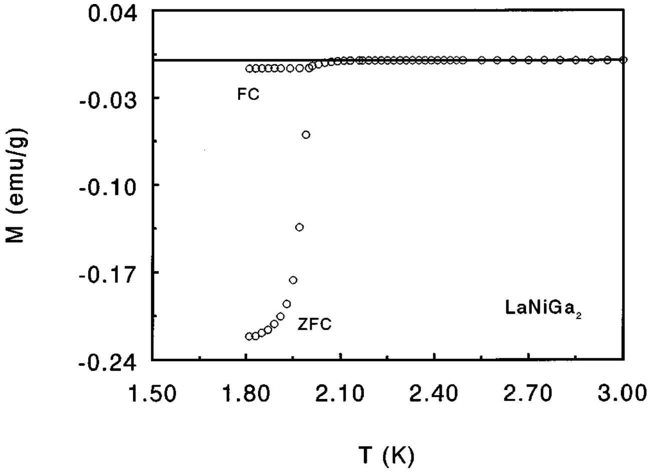

FIG. 2. Temperature dependence of the zero-field cooled (ZFC) and field-cooled (FC) magnetization data for the compound $LaNiGa_2$ measured in a field of $H=10$ Oe between 1.80 and 3.0 K.

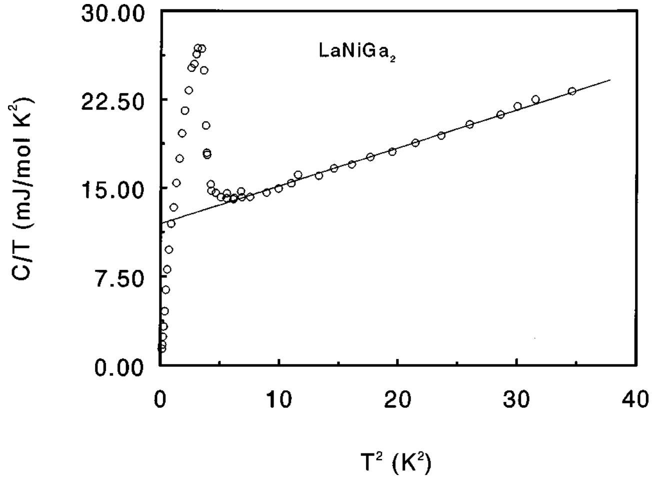

FIG. 4. Specific heat divided by temperature C/T vs T^2 of $LaNiGa_2$ between 0.35 and 6.0 K. The value of γ was obtained by extrapolating the specific heat in this plot of C/T vs T^2 down to 0 K.

Earth's magnetic field. The sample was attached to a sapphire chip, which has two separated silicon films deposited on it to serve as a heater and a thermometer. The calibration of the thermometer was done against a calibrated germanium thermometer. For each point of the specific-heat measurements, a small heat power was introduced to the chip and the thermal relaxation was measured and analyzed to obtain the specific heat of the sample. Figure 3 displays the temperature dependence of the specific C for the compound $LaNiGa_2$ between 0.35 and 6 K. In both χ_{dc} and C measurements the transition occurs over an identical interval of about 0.08 K (10–90 % values). The agreement of transition temperatures measured by both techniques are clear evidence of bulk superconductivity in $LaNiGa_2$. The specific-heat data plotted as C/T against T^2 in Fig. 4 show that the heat capacity C of $LaNiGa_2$, in the normal state at temperatures below 6 K, can be fitted to the expression $C_n=\gamma T+\beta T^3$ by a least-squares analysis, which yields the value $\gamma=11.64\pm0.05$ mJ/mol K^2 and $\beta=0.332\pm0.006$ mJ/mol K^4, the latter value corresponding to the Debye temperature $\Theta_D=560\pm10$ K. For comparison to γ, we have estimated γ_{BCS} from the measured specific heat jump ΔC (32 ± 2 mJ/mol K) at T_c (1.97 ± 0.02 K) using the Bardeen-Cooper-Schrieffer (BCS) relation $\gamma_{BCS}=\Delta C/1.43T_c\sim11.36$ mJ/mol K^2. The observed value of γ (11.64 ± 0.05 mJ/mol K^2) is in good agreement. According to the microscopic theory of superconductivity, the specific-heat data below T_c have a dominant low-temperature behavior of the form $\exp[-\Delta(0)/k_BT_c]$. Analysis of our data shown in Fig. 3, that is, $2\Delta(0)=3.2k_BT_c$, provides evidence that the gap is of the order of 5.2 meV.

dc electrical resistivity measurements were made between 1.80 and 300 K using a standard four-probe technique in a Quantum Design system fully automated for temperature stability and data acquisition.[12] Fine platinum wires (~2 mil diameter) were spot welded to the rectangular-shaped sample, and served as the voltage and current leads. The complete resistivity data between 1.80 and 300 K for the polycrystalline sample $LaNiGa_2$ are presented in Fig. 5. It is seen that the residual resistivity and residual resistivity ratio values for superconducting $LaNiGa_2$ are 14.1 $\mu\Omega$ cm and 5.2, respectively. The zero resistance temperature (2.05 K),

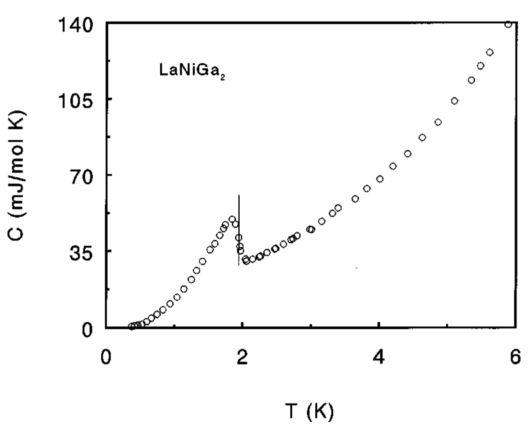

FIG. 3. Heat capacity of specimen $LaNiGa_2$ between 0.35 and 3.0 K in the Earth's field.

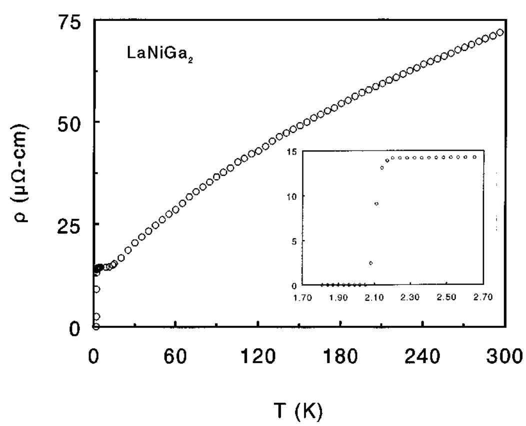

FIG. 5. Electrical resistivity vs temperature between 1.80 and 300 K for $LaNiGa_2$. Inset: ρ vs T between 1.80 and 2.70 K.

shown in the inset of Fig. 5, is slightly higher than the transition point value obtained by dc susceptibility and heat-capacity measurements. This phenomenon is probably due to the surface superconductivity[13,14] of this compound. As a concluding remark, the Ni-based intermetallic compound $LaNiGa_2$, which crystalizes in the $NdNiGa_2$-type structure, exhibits phonon-mediated superconductivity with $T_c \sim 1.97 \pm 0.02$ K, as characterized by the magnetic, specific-heat, and resistivity data.

We are grateful to Dr. Y. Y. Chen for assistance in using the calorimeter. Research at Chung Cheng University was supported by the National Science Council of the Republic of China under Contract No. NSC90-2112-M194-015.

*Email address: whlee@phy.ccu.edu.tw

[1] H. Rosner, R. Weht, M. D. Johannes, W. E. Pickett, and E. Tosatti, Phys. Rev. Lett. **88**, 027001 (2002).
[2] T. He, Q. Huang, A. P. Ramirez, Y. Wang, K. A. Regan, N. Rogado, M. A. Hayward, M. K. Haas, J. S. Slusky, K. Inumaru, H. W. Zandbergen, N. P. Ong, and R. J. Cava, Nature (London) **441**, 54 (2001).
[3] W. H. Lee and H. K. Zeng, Solid State Commun. **101**, 323 (1997).
[4] W. H. Lee, H. K. Zeng, Y. D. Yao, and Y. Y. Chen, Physica C **266**, 138 (1996).
[5] W. H. Lee, F. A. Yang, C. R. Shih, and H. D. Yang, Phys. Rev. B **50**, 6523 (1994).
[6] Chandan Mazumdar, R. Nagarajan, C. Godart, L. C. Gupta, M. Latroche, S. K. Dhar, C. Levy-Clement, B. D. Padalia, and R. Vijayaraghavan, Solid State Commun. **87**, 413 (1993).
[7] R. Nagarajan, Chandan Mazumdar, Zakir Hossain, S. K. Dhar, K. V. Gopalakrishnan, L. C. Gupta, C. Godart, B. D. Padalia, and R. Vijayaraghavan, Phys. Rev. Lett. **72**, 274 (1994).
[8] R. J. Cava, H. Takagi, H. W. Zandbergen, J. J. Krajewski, W. F. Peck Jr, T. Siegrist, B. Batlogg, R. B. van Dover, R. J. Felder, K. Mizuhashi, J. O. Lee, H. Eisaki, and S. Uchida, Nature (London) **367**, 252 (1994).
[9] T. Siegrist, H. W. Zandbergen, R. J. Cava, J. J. Krajewski, and W. F. Peck Jr., Nature (London) **367**, 254 (1994).
[10] Yu. M. Grin' and Ya. P. Yarmolyuk, Dopov. Akad. Nauk. Ukr. RSR, Ser. A: Fiz.-Mat. Tekh. Nauki **44**, 69 (1982).
[11] R. Bachmann, F. J. Disalvo, T. H. Geballe, R. L. Greene, R. E. Howard, C. N. King, H. C. Kirsch, K. N. Lee, R. E. Schwall, H. U. Thomasand, and R. B. Zubeck, Rev. Sci. Instrum. **43**, 205 (1972).
[12] Quantum Design, Inc., San Diego, CA.
[13] D. Saint James and P. G. de Gennes, Phys. Lett. **7**, 306 (1963).
[14] J. G. Park, Phys. Rev. Lett. **16**, 1196 (1966).