Hall mobility of positive carriers in the semimetallic compound LiGa

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(Received 6 October 1988; revised manuscript received 20 March 1989)

The temperature-dependent Hall coefficient and Hall mobility above 90 K of LiGa are presented for the Li-deficient phase. These results indicate that the holes are the majority carriers in LiGa and their mobility is dominated by the lattice-phonon scattering. The difference in the hole mobility between LiGa and LiAl suggests that the band overlap of the hole and electron pockets is greater in LiGa than in LiAl. The "positive" Hall coefficient is also associated with the lowering of the Fermi energy in the "real" defect phase LiGa resulting from the constitutional lithium vacancies.

We have reported[1] that the electrical transport properties of LiGa, with holes as the majority carriers, correlate with the semimetallic band structure calculated by Schmit.[2] Furthermore, we have found an abrupt change near 230 K in the electrical resistivity[1] that occurs near the phase boundary of the Li-deficient region in LiGa. As a result, we have proposed that these anomalies are associated with the low-temperature ordering of Li vacancies. LiGa [NaTl structure,[3] space group $Fd3m$ (O_h^7)], is composed of two interpenetrating sublattices, each forming a diamond lattice. The LiGa compound has a relatively high melting point (~740 °C),[4] a well-defined structure, and a homogeneity range of approximately 46–54 at. % Li.[4,5]

In this paper, we report measurements of the temperature-dependent Hall coefficient (R_H) and Hall mobility above 90 K and discuss the origin of the scattering center for holes.

LiGa crystals were grown by the same method[6] used for LiAl single-crystal growth with pyrolytic boron nitride crucibles. As-grown crystals, with ~1 cm diameter, contained several grain boundaries. Typical samples used in this investigation were Li-deficient (46.5±0.5 at. % Li) LiGa. Temperature-dependent Hall-effect measurements were made using a modified ac Hall-effect measuring system[7] designed to diminish or eliminate the Ettingshausen effect with its accompanying unwanted thermal electromotive force. This method makes use of a constant magnetic field (7.5 kG) and alternating driving currents. In order to minimize the generation of an unbalanced voltage, square-shaped slabs were used as the samples. Also, the voltage and current probes were located exactly on the corners of the sample. An alternating current of 500 mA, which is amplified by an audio amplifier, goes into the sample through an isolation transformer. The low-level Hall voltage, over the range 10–100 nV, was amplified by a lock-in amplifier (PAR model HR-8) through a preamplifier. Most measurements were carried out at frequencies ranging from 10 to 70 Hz—high enough to use a conventional audio-frequency transformer, and low enough to avoid the skin effect in samples with metallic conductivity.

The temperature-dependent electrical resistivity for Li-deficient LiGa (46.5 at. % Li) is given in Fig. 1. This figure shows that an anomaly[1] occurs near the critical temperature (T_c = 230 K) in the resistivity curve. The curve is characterized by a monotonic increase with increasing temperature—except for the anomalous part. The variation of the Hall coefficient with temperature is shown in Fig. 2. The Hall coefficient in LiGa shows a "positive" value similar to that seen in β-LiAl.[1,8,9] There is a slight scatter in R_H values. The R_H curve increases slightly as the temperature is decreased but it does not undergo any significant change at T_c. Furthermore, the R_H values [$(2.5-3)\times10^{-4}$ cm^3/C] of LiGa are lower than those of β-LiAl (~2.5×10^{-3} cm^3/C) at the Li-

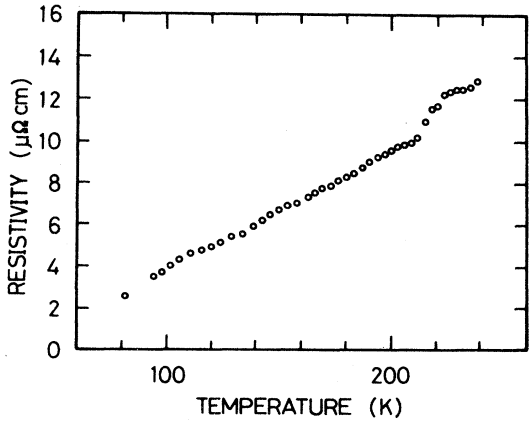

FIG. 1. Temperature-dependent electrical resistivity of Li-deficient LiGa (46.5 at. % Li). The size of the data points is the estimated error.

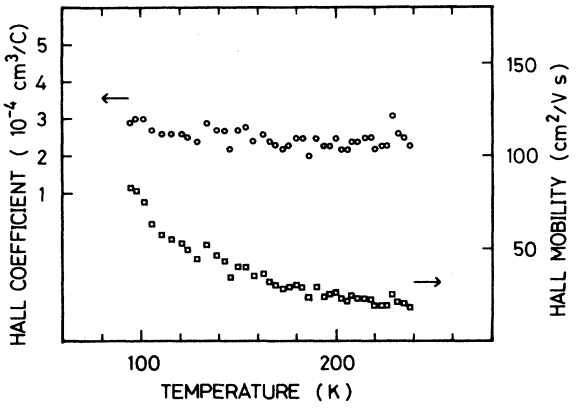

FIG. 2. Hall coefficient and Hall mobility of LiGa (46.5 at. % Li) as a function of temperature. The estimated error is ±3%.

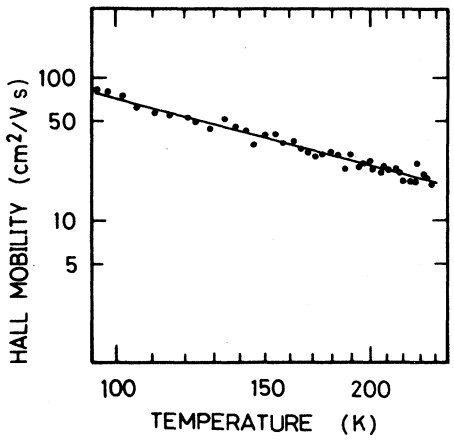

FIG. 3. $\ln(\mu_H)$ vs $\ln(T)$ plot for LiGa (46.5 at. % Li). The exponent of T was determined to be -1.54, indicating lattice-phonon scattering.

deficient phase boundary.[8] Although the electronic structure of LiGa has not been studied extensively, it is expected that the energy-band structure of LiGa is similar to that of β-LiAl.[10,11] A recent calculation indicates that LiGa is a semimetallic compound with a hole pocket around point Γ and an electron pocket along the $\Gamma-X$ direction and around point L of the Brillouin zone.[2] The band overlap of the hole and electron pockets is presumably greater in LiGa than in LiAl. Our small positive values of R_H are consistent with the calculations. The band structure for the "real" defect phase LiGa shows a lowering of the Fermi energy[11] due to the constitutional lithium vacancies.[1,12] As a result of this shift, the large hole pocket surrounding Γ overwhelms the small electron pockets along $\Gamma-X$ and around L.

Figure 2 also shows the temperature-dependent Hall mobility ($\mu_H=R_H/\rho$). The Hall mobility decreases almost exponentially with increasing temperature. In order to survey the origin of the scattering center for holes near the Li-deficient phase boundary, the $\ln(\mu_H)$ versus $\ln(T)$ curve is plotted in Fig. 3. The Hall mobility can be fairly well represented by a power-law dependence on the temperature, with the power of -1.54. This value is smaller than the exponent of T for scattering between carriers in different valleys observed in bismuth.[13] In either the electron-hole or the anisotropic-carrier case, the temperature dependence is T^{-2}. On the other hand, the temperature dependence of the mobility associated with lattice scattering follows approximately the $T^{-3/2}$ law predicted by simple theory.[14] Therefore, the hole mobility near the Li-deficient phase boundary above 90 K essentially might be dominated by the lattice-phonon scattering rather than the ionized-impurity scattering based on the field of a Li vacancy.[1,12] The room-temperature hole mobility for Li-deficient LiGa is ~25 cm^2/V s, while that of LiAl (Refs. 7 and 9) is 60–70 cm^2/V s. The difference in the hole mobility between LiGa and LiAl must be based on the degree of band overlap of the hole and electron pockets as discussed above.

In conclusion, we find that the holes are the majority carriers in LiGa and their mobility is dominated by the lattice-phonon scattering above 90 K. We propose that the band structure for the "real" defect phase LiGa indicates a lowering of the Fermi energy resulting from the constitutional lithium vacancies. The "positive" Hall coefficient indicates that the large hole pocket surrounding Γ overwhelms the small electron pockets along $\Gamma-X$ and around L of the Brillouin zone.

This work was supported in part by the Division of Materials Sciences, Office of Basic Energy Science, U.S. Department of Energy, under Contract No. W-31-109-ENG-38.

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