

E_g modes and F_{2g} modes, respectively. The value of B which we obtain for Li^+ in KCl by the method of Sec. IV (see Table V) yields resonance modes of symmetry E_g at the following frequencies: $\omega_1 \sim 47 \text{ cm}^{-1}$, $\omega_2 \sim 76 \text{ cm}^{-1}$, $\omega_3 \sim 101 \text{ cm}^{-1}$, and $\omega_4 \sim 111 \text{ cm}^{-1}$. Observed spectra² of Li^+ in KCl exhibit peaks at 130 cm^{-1} and 200 cm^{-1} which might correspond to the second-order processes $\omega_1 + \omega_2$ and $2\omega_3$. However, such comparison with experiment is not very meaningful in the present circumstances due to the approximations made in the theory, viz., the restriction to nearest-neighbor interactions only, the lattice dynamical model used to represent the perfect crystal, and its use of 0°K data. All these factors contribute to a shift in the resonant-mode frequency. Also, no assignment of the peaks in

the observed spectra² to modes of particular symmetry has been made.

It is to be hoped that there will soon be sufficient experimental data available to obtain a set of values for the parameters which enter a calculation of the impurity induced first-order Raman and infrared spectra of alkali halide crystals. The consistency of these values would provide a test of the lattice dynamical model used in the calculations.

ACKNOWLEDGMENT

It is a pleasure to thank Professor A. A. Maradudin for many helpful discussions during the course of this work.

Resistance and Magnetoresistance of $\text{Li}(\text{NH}_3)_4^\dagger$

W. J. McDONALD* AND J. C. THOMPSON

The University of Texas, Austin, Texas

(Received 22 March 1966)

Measurements are reported on the resistivity and magnetoresistance of the bronze-colored compound formed on freezing a saturated solution of lithium in ammonia. The lithium compound freezes at 89°K . The residual resistivity at 4.2°K is $0.30 \mu\Omega \text{ cm}$. The lithium compound shows a nonsaturating magnetoresistance of the form $\Delta\rho/\rho_0 = AB^2$ where $A = 46 \text{ m}^4 \text{ volt}^{-2} \text{ sec}^{-2}$, at fields up to 43 kG. The large magnetoresistance is assumed to originate from hole and electron bands containing equal numbers of carriers; that is, the material is taken to be completely compensated.

INTRODUCTION

THE increased interest in liquid metals in the last decade has stimulated interest in solutions of alkali metals and alkaline earth metals in ammonia as they exhibit metallic behavior. At least two of the metals in group I and II elements (all of which are soluble in liquid ammonia) form solid compounds with the ammonia; they are lithium and calcium.¹ These compounds have the approximate formulas $\text{Li}(\text{NH}_3)_4$ and $\text{Ca}(\text{NH}_3)_6$. We report here measurements on the lithium compound: lithium tetrammine.

As this material has not been extensively studied, a few qualitative observations are in order. The solid is a golden-red color, the same as the liquid. No sign is seen of metal precipitation from the solution on freezing, as with Na-NH_3 . There is no large change in volume on freezing, so the density must be close to that of the liquid (0.5 g/cm^3). The crystal structure is not known

for $\text{Li}(\text{NH}_3)_4$: for $\text{Ca}(\text{NH}_3)_6$ Cagle reports² body-centered cubic. The only other previous work is that of Jaffe,³ Birch and MacDonald,⁴ and Morgan *et al.*,⁵ all on the lithium compound. They reported, respectively, Hall data and magnetoresistance, freezing points and composition, and resistivity and several phase changes. The freezing point at 89°K has the appearance of a eutectic; it lies at a minimum in a melting point-versus-composition diagram. Hence 89°K may not be the melting point of $\text{Li}(\text{NH}_3)_4$, as melting points of compounds are usually found to be at maxima. However, the character of the material in the solid does not fit ideas of either a solid solution or a metal precipitated from the solvent. Furthermore, examples can be found wherein "submerged maxima" or peritectics lie beneath, or nearly beneath, eutectics.⁶ Hansen⁷ reports such struc-

[†] Research assisted by the U. S. Office of Naval Research and the R. A. Welch Foundation.

* Texas Instruments Fellow in Physics. Present address: Esso Production Research Company, Houston, Texas 77001.

¹ M. J. Sienko, in *Metal-Ammonia Solutions*, edited by G. Lepoutre and M. J. Sienko (W. A. Benjamin, Inc., New York, 1964), p. 23.

² H. J. Holland and F. W. Cagle, 145th National Meeting American Chemical Society, 1963 (unpublished).

³ H. Jaffe, *Z. Physik* **93**, 741 (1935).

⁴ A. J. Birch and D. K. C. MacDonald, *Trans. Faraday Soc.* **43**, 792 (1947); **44**, 735 (1948).

⁵ J. A. Morgan, R. L. Schroeder, and J. C. Thompson, *J. Chem. Phys.* **43**, 4494 (1965).

⁶ Eric H. Bucknall (private communication).

⁷ M. Hansen, *Constitution of Binary Alloys* (McGraw-Hill Book Company, Inc., New York, 1958), 2nd ed., pp. 631, 905, 1058.

ture for CuSi alloys (near 20% Cu), LiTl alloys (near 34% Tl), and NiW alloys (near 20% W).

We report here the results of electrical resistivity measurements on this unusual compound. The material is metallic in behavior, having a low residual resistance and a resistivity which increases with increasing temperature. Also, we report the observation of a large, nonsaturating magnetoresistance in lithium tetrammine in magnetic fields to 43 kG. A large magnetoresistance is usually associated with a long electron mean free path and either a highly distorted Fermi surface with open orbits or a completely compensated material with equal numbers of holes and electrons, and has been observed heretofore only in very pure metallic single crystals.

EXPERIMENTAL

Samples were prepared by distilling NH_3 onto lithium at 200°K until a solution of the desired concentration was produced, then siphoning the solution into a cylindrical quartz cell 0.7 cm i.d. \times 10 cm long. The solution was then immersed in liquid nitrogen while the cell was sealed off. Conductivity measurements were made over long periods and after repeated melting-freezing cycling. No variation with time was observed so long as the solid was allowed to anneal for a few hours, as described below.

$\text{Li}(\text{NH}_3)_4$ is a reactive and unstable material which decomposes to a metal amide plus hydrogen quite readily.⁵ This decomposition is catalyzed by the presence of water, other impurities such as oxides of the alkali metal used, and the usual electrode materials: platinum and tungsten. A problem encountered in the solid phase is that crystallites may form and separate as the solution is frozen. This may cause a significant variation from run to run in the form factor necessary for calculating the conductivity from the usual four-point probe method. For these reasons, it was decided to use an electrodeless technique such as that described by Zimmerman⁸ and others^{9,10} for measuring the electrical conductivity of this material. Only quartz or glass comes in contact with the sample. This method has the additional advantage that it measures the bulk conductivity and possible inhomogeneities in the materials will tend to average out.

The technique employed is essentially that of measuring the change in Q of a coil when a cylindrical sample is introduced into its core. Eddy current losses in the core effect this change in the Q of the coil. The quantity actually measured is either the change in resistance ΔR or the change in inductance ΔL of the coil on insertion of the sample into the coil. The equations necessary for calculation of the conductivity from either of these

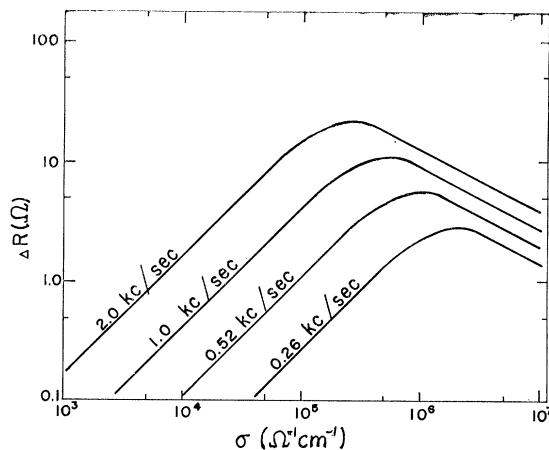


FIG. 1. Computed curves relating the measured resistance change ΔR to the conductivity of the sample for several frequencies. The maximum in each curve occurs at conductivities for which the skin effect becomes appreciable.

quantities are given by MacLachlan¹¹

$$\alpha \frac{\Delta L}{L_0} = \frac{2M_1(x)}{xM_0(x)} \cos[\theta_1(x) - \theta_0(x) - \frac{3}{4}\pi] - 1, \quad (1)$$

$$\alpha \frac{\Delta R}{\omega L_0} = \frac{2M_1(x)}{xM_0(x)} \sin[\theta_1(x) - \theta_0(x) - \frac{3}{4}\pi], \quad (2)$$

where $x^2 = \omega\mu a^2\sigma$, $M(x)$, $\theta_1(x)$, and $\theta_2(x)$ are spherical Bessel functions, ω is the frequency of operation, a is the sample radius, σ is the conductivity, μ is the permeability (taken as unity), and α , the constant of proportionality, is equal to the coil to sample cross-sectional area ratio.

Because the change in R produced by a sample of given conductivity is about one hundred times as large as the corresponding change in L for our arrangement, R is the quantity most convenient with which to work. It is also convenient to work in a region where all but the first term of the expansion of Eq. (2) may be neglected and the conductivity is linearly dependent on ΔR . When this is not possible it is possible to solve the equation for conductivity in terms of ΔR on a computer: typical curves appear in Fig. 1.

Figure 2 is a block diagram of the circuit used to detect the resistance changes. The detection coil formed one leg of a series resonant bridge, the remainder of which was constructed from components purchased from the General Radio Company and assembled according to their instructions.¹²

Figure 3 is a drawing of the experimental apparatus. Capital letters refer to that drawing. The coil mount was constructed of thin-wall stainless tubing, A . Sol-

⁸ J. E. Zimmerman, Rev. Sci. Instr. **32**, 402 (1961).

⁹ C. P. Bean *et al.*, J. Appl. Phys. **30**, 1976 (1959).

¹⁰ D. W. Nyberg and R. E. Burgess, Can. J. Phys. **40**, 1174 (1962).

¹¹ N. W. MacLachlan, *Bessel Functions for Engineers* (Oxford University Press, Oxford, England, 1934).

¹² I. G. Easton, General Radio Company Cambridge, Massachusetts (unpublished report).

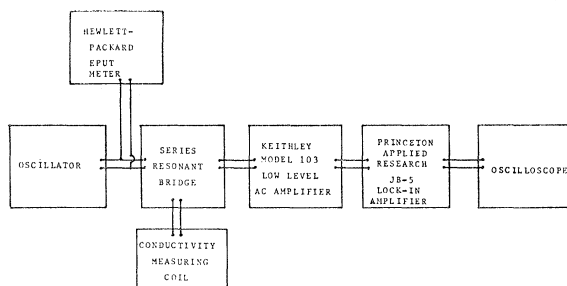


FIG. 2. Block diagram of the ac conductivity-measuring circuit.

dered to the lower end of this tube was a section of copper tubing, B. The solenoid C, thermometers D, and heater E, were wound on a Bakelite form, 2 in. long. The Bakelite form was epoxied to the lower end of the copper tubing. The upper end of the stainless tubing was soldered into a mounting flange G and a manifold which provided access for the vacuum system, electrical leads, and sample. The top of the manifold H was removable to allow the sample to be introduced into the cryostat. The samples, sealed in quartz cells, were fitted into stainless-steel clamps on the end of a thin-walled stainless-steel tube, which was sufficiently long to reach from the top of the manifold to the coil. A 6-in. section of brass rod was soldered on top of this. This brass rod fitted through a hole with an "O" ring seal in the center of the top plate of the manifold. Thus the sample could be moved in and out of the coil by raising and lowering the brass rod, even when the system was evacuated as necessary for thermal isolation. Finally, a vacuum jacket of thin-walled stainless-steel tubing was fitted over the apparatus and sealed by means of an "O" ring to the under side of the mounting flange.

The coil for the ac conductivity measuring system consisted of 3500 turns of No. 40 AWG Nyclad covered wire, wound in eight layers on a Bakelite form of 0.972 cm i.d. The coil was 4 cm long and had a mean diameter of 1.15 cm. The inductance of the coil was 12.1 mH and it had a room-temperature resistance of 171 Ω .

Because of its wide useful temperature range and reproducibility, copper-resistance thermometry was used. The thermometer consisted of two bifilarly wound coils connected in series; each coil was wound of No. 40 AWG copper wire and was mounted directly on the conductivity coil form one at each end of the conductivity coil. The thermometer was calibrated at liquid-helium, liquid-nitrogen, ice-water, and room-temperature points. The calibration matched the table of Dauphinee and Preston-Thomas¹⁸ nicely. This table was then used for temperature determination. The calibration was checked at the nitrogen boiling point often and was found not to vary.

A platinum-resistance thermometer calibrated by the National Bureau of Standards was used in the 14 to

30°K range. This was necessary for obtaining temperature measurements below 25°K, where the copper-resistance thermometers begin to lose sensitivity. The platinum thermometer was mounted parallel to and within $\frac{1}{4}$ in. of the coil; the resistance measuring circuit for it was identical with that of the copper thermometer.

To make a measurement, the desired frequency of operation (usually in the range of 200 to 5000 Hz) was set using an electronic counter; the lock-in amplifier was tuned and the series resonant bridge balanced, first without the sample and then with the sample in place. The phase-sensitive detection of the lock-in amplifier allowed the R and L nulls to be detected independently since ΔR and ΔL affect the in-phase and out-of-phase voltage, respectively. ΔR was determined from the two balance conditions (sample in, sample out) and the conductivity read from curves such as those in Fig. 1. The accuracy of the resistivity measuring apparatus was checked using copper cylinders and liquid mercury as samples.

The sample temperature was changed by varying the bath temperatures whenever possible; for temperatures not accessible in this way the sample chamber was evacuated and the heater used to raise the temperature. The temperature range above the helium boiling point and below 30°K proved inaccessible to measurements of conductivity with this arrangement because of problems of temperature control. Because this was a temperature range of great interest, a run was made in the experimental chamber of an ADL Collins helium liquifier with the temperature being stabilized at many points between 14 and 38°K by the experienced operator of this machine.

Magnetoresistance data were taken only in the liquid baths at 4.2 and 77.4°K. Two different field configurations were employed. For low-field data ($B \leq 7$ kG) the field was perpendicular to the coil (and sample) axis "transverse." The eddy-current loops produced in the

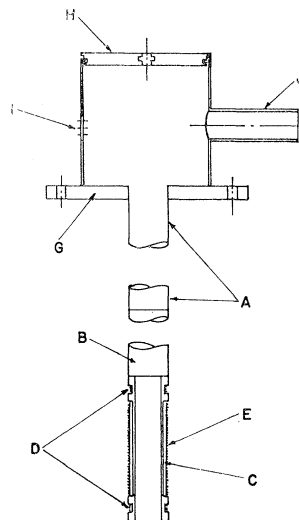


FIG. 3. Schematic of the sample holder. The sample is located in the coil C. The other labels are identified in the text.

¹⁸ T. M. Dauphinee and H. Preston-Thomas, Rev. Sci. Instr. 25, 884 (1954).

sample by the coil would be coaxial with coil and sample in the absence of a field. The "transverse" field would produce a couple tending to rotate the loops to a position coaxial with the field lines. Though a detailed analysis has not been made we presume this means that field and current are perpendicular as in conventional measurements of transverse magnetoresistance. This impression is reinforced by measurements made in a "longitudinal" geometry where the field is parallel to the cylinder axis ($B \leq 43$ kG). Here the external field is parallel to the field produced by the coil, hence perpendicular to the eddy-current loops. The results were identical for the two field orientations and are taken to be characteristic of transverse magnetoresistance. The magnetic field was varied in steps of approximately 250 G for the "transverse" field and 1000 G for the "longitudinal" field.¹⁴ Otherwise magnetic field runs were identical to those with no magnetic field.

After each run the sample was immediately replaced in a Dewar of liquid nitrogen for storage.

RESULTS AND DISCUSSION

The results of the experimental determination of the electrical conductivity and the magnetoresistance of $\text{Li}(\text{NH}_3)_4$ are presented in the following figures. The relative accuracy of the experimental points is better than $\pm 1\%$ with both the lithium and the calcium compounds over the entire temperature range, with the exception of the zero-magnetic-field data in the liquid-helium region. Accuracy here was limited to $\pm 10\%$. The reduced accuracy occurred because it was necessary to make measurements in this temperature range at fre-

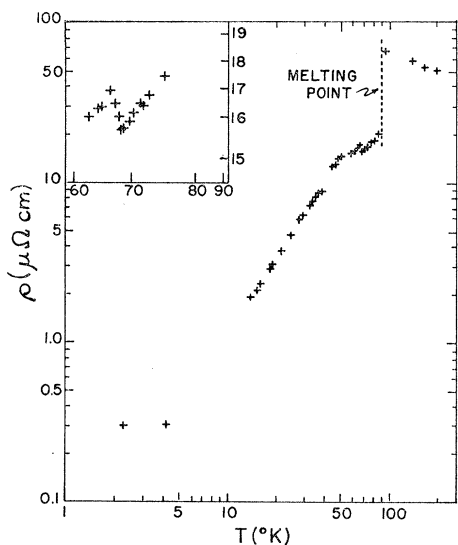


FIG. 4. Resistivity versus temperature for $\text{Li}(\text{NH}_3)_4$. The inset shows the discontinuity at 67°K in detail.

¹⁴ High-field data were taken in a Westinghouse superconducting solenoid at the Southwest Research Institute, San Antonio with the cooperation of F. L. Whitmore.

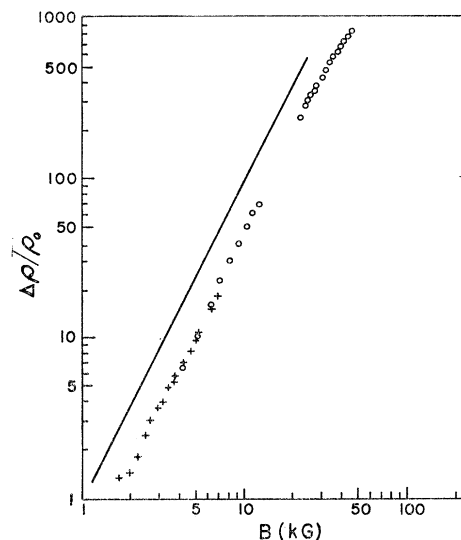


FIG. 5. Magnetoresistance of $\text{Li}(\text{NH}_3)_4$ at 4.2°K. The solid line indicates a proportionality to B^2 . The + indicate field perpendicular to coil, the o indicate B parallel to the coil.

quencies where the skin depth is less than the sample radius. The accuracy with which the absolute value of the resistivity is known is approximately $\pm 20\%$. This limitation is due to uncertainty in the fill factor α and in the radius of the sample.

The electrical-conductivity data for the tetrammine lithium are presented in Fig. 4. The curve represents the result of measurements made on two different samples totaling over 200 data points. The immediate and obvious result of this experiment is that $\text{Li}(\text{NH}_3)_4$ may be classed as a metallic conductor. This is in agreement with the conclusions drawn from Hall measurements on the liquid as well as with the measurements of Morgan *et al.* in the liquid-nitrogen temperature range. The residual resistivity of $0.30 \mu\Omega\text{cm}$ reported here is appropriate for a metal, though it is not so low as that for the "good" conductors.

A comparison with pure lithium is interesting. Pure polycrystalline lithium has a residual resistivity of $0.05 \mu\Omega\text{cm}$, according to MacDonald *et al.*¹⁵ On an atomic basis, the $\text{Li}(\text{NH}_3)_4$ contains only one-fifth the lithium as the pure metal; the ratio of the conductivities is nearly the same. As noted by Jaffe, the ratios also hold if the conductivities are determined at the respective melting points. It must be emphasized that the material discussed here is not a solid solution of lithium in ammonia but a compound. As the density of $\text{Li}(\text{NH}_3)_4$ is approximately the same as that of Li,¹⁶ the number of lithium atoms per unit volume is ten times larger in the pure metal than in the tetrammine. Were one to treat the lithium tetrammine as a simple monovalent metal

¹⁵ D. K. C. MacDonald, G. K. White, and S. B. Woods, Proc. Roy. Soc. (London) A235, 358 (1956).

¹⁶ The density of the solid is close to that of the liquid, which is close to that of pure Li.

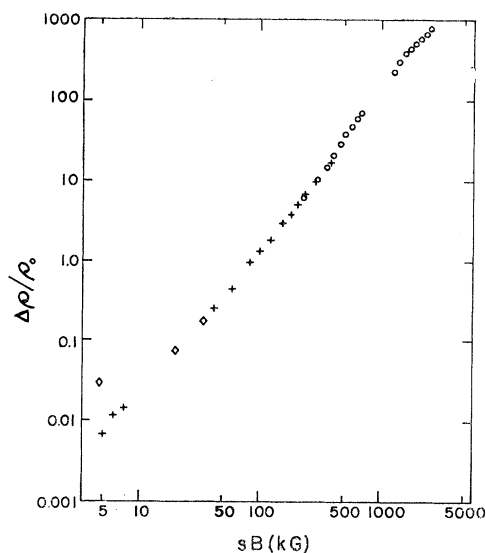


FIG. 6. A reduced Kohler plot. $s = \rho_0(77^\circ\text{K})/\rho_0(T)$. At 4.2°K , $s = 60$. The $+$ and \circ have the same meaning as in Fig. 5; the \diamond are data from Ref. 3.

(which we do not) the preceding numbers imply a higher electron mobility in the compound than in the pure metal. The magnetoresistance measurements discussed in the next section oblige us to adopt a less naive view of the compound insofar as the electron density is concerned.

The effect of temperature on resistivity in the liquid-nitrogen temperature range has been studied by Morgan *et al.*⁵ for $\text{Li}(\text{NH}_3)_4$. They observe a discontinuity at 83°K as well as the break near 69°K . Their samples, more carefully annealed than those used here, showed a higher conductivity near the melting point but are in essential agreement with the present results elsewhere. Below 69°K the resistivity is at first a linear function of T , increasing finally to $T^{1.5}$ over a large portion of the low-temperature range. The limiting value of the resistivity is reached near 10°K . The low temperature at which impurity limitation of the conductivity appears is apparently the result of a low Debye temperature, rather than a low level of impurities.

Figure 5 shows the extraordinarily large [$(\Delta\rho/\rho_0) = 850$ at 43 kG] magnetoresistance exhibited by the solid $\text{Li}(\text{NH}_3)_4$ at liquid-helium temperatures. The apparent magnitude of the magnetoresistance casts some doubts on the effect of galvanomagnetic terms on the conductivity measurement; these drop out. The magnetoresistance of copper was measured at 77°K , where it has approximately the same zero-field resistivity as the $\text{Li}(\text{NH}_3)_4$ at 40°K ; no unusual behavior of the system was observed. Also the magnetoresistance of bismuth was measured at liquid-nitrogen temperature with the apparatus, and the results agreed well with published data. These and other attempts to attribute the results to the ac measuring system, either as in-phase pick-up

or a conceptual problem, were not successful, and we conclude that the magnetoresistance is indeed real.

It was shown by Kohler¹⁷ that the dependence of the magnetoresistance on temperature, magnetic field, and sample purity should be a function of τB only. Since τ is proportional to $1/\rho$, one usually writes Kohler's rule in the form

$$\Delta\rho/\rho_0 = f(B/\rho_0). \quad (3)$$

One plots $(\Delta\rho/\rho_0)$ versus (B/ρ_r) , where ρ_r is the ratio of the zero-field resistivity at the temperature where the measurement is made to the zero-field resistivity at some reference temperature. For this type of plot, measurements of magnetoresistance at different temperatures and on samples of different purity should all fall on the same curve. In order to compare various materials on a Kohler plot, a standard reference temperature is necessary; this is usually taken to be the Debye temperature. If one is only examining the consistency of magnetoresistance data for a single material, an arbitrary reference temperature will suffice. We use 77°K .

Figure 6 is a reduced Kohler plot for the solid $\text{Li}(\text{NH}_3)_4$. Included on this curve are the three measurements of $(\Delta\rho/\rho_0)$ made by Jaffe³ in the liquid-hydrogen temperature range. With the exception of the lowest of these (highest temperature), his data are in good agreement with those obtained here at 77°K and 4.2°K .

From Fig. 5 we see that within experimental error the magnetoresistance is quadratically dependent on the field and shows no tendency to saturate even at fields of 43 kG. No oscillatory behavior was observed.

According to the Frank formula¹⁷ the change in resistivity is given by

$$\Delta\rho/\rho_0 = AB^2/(1 + CB^2), \quad (4)$$

where C is the square of the mobility.

This reduces in the present case to

$$\Delta\rho/\rho_0 = AB^2, \quad (5)$$

since, over the range of fields covered, there was no departure from a quadratic dependence. Solving for the coefficient of magnetoresistance A one obtains

$$A = 46 \text{ (m}^2/\text{V sec)}^2.$$

Since there was no tendency to saturation, it is impossible to solve the Frank equation for the mobility. The value of the mobility μ calculated from the usual expression $\mu = \sigma/n_0e$ is $0.5 \text{ m}^2/\text{V sec}$, with $n_0 = 4 \times 10^{21} \text{ cm}^{-3}$.¹⁶

Jan¹⁷ shows that it is convenient to describe the anisotropy of the Fermi surface in a one-band model in terms of a dimensionless quantity Q defined as

$$Q^2 = \frac{\langle f^2\tau^3 \rangle_{\text{av}} \tau - \langle f\tau^2 \rangle_{\text{av}}^2}{\langle \tau^4 \rangle_{\text{av}}}, \quad (6)$$

¹⁷ J. P. Jan, in *Solid State Physics* (Academic Press Inc., New York, 1957), Vol. V, p. 1.

where τ is the relaxation time and $f=m/m^*$; both τ and f are taken to be wave-number-dependent. For a transverse magnetic field,

$$\Delta\rho/\rho_0=\mu^2Q^2B^2. \quad (7)$$

Further interpretation of the magnetoresistance data is at best complex. The two-band model¹⁸ predicts that for equal numbers of holes and electrons, the transverse magnetoresistance will not saturate. This is what was observed in the $\text{Li}(\text{NH}_3)_4$; the behavior of the lithium tetrammine appears similar to that of completely compensated metals such as molybdenum or tungsten as described by Fawcett and Reed.¹⁹ A completely compensated metal is one in which the electron and hole densities are equal and the Fermi surfaces, for both holes and electrons are closed. The model is usually applicable only to divalent metals; however, if the phase transformation at 69°K is to the hcp structure one could have two electrons per unit cell. For a compensated metal on the two-band model, if holes and electrons are assumed to have both equal densities and equal mobilities, the Q of Eq. (7) may be replaced by unity, therefore μ^2 is equal to the coefficient of magnetoresistance, A :

$$\mu^2=46 \text{ (m}^2/\text{V sec)}^2, \quad (8)$$

or

$$\mu=6.8 \text{ (m}^2/\text{V sec)},$$

more than an order of magnitude larger than the mobility calculated from the conductivity. This suggests that although the hole and electron densities may be the same, their mobilities are not. For unequal mobilities¹⁶ one obtains

$$\Delta\rho/\rho_0=\mu_e\mu_hB^2. \quad (9)$$

Thus

$$\begin{aligned} \mu_e\mu_h &= \Delta\rho/\rho_0/B^2 \\ &= A, \text{ the coefficient of magnetoresistance} \\ &= 46 \text{ m}^2/\text{V sec}. \end{aligned}$$

One must now interpret the conductivity on a two-band model. The total conductivity is the sum of the conductivities of each of the carriers

$$\begin{aligned} \sigma &= \sigma_e + \sigma_h, \\ \mu &= \mu_e + \mu_h. \end{aligned} \quad (10)$$

Up to now we have taken $\mu=0.5 \text{ m}^2/\text{V sec}$. In obtaining this result we assumed that each lithium atom contributed one carrier, an electron. On the two-band scheme the carrier density is less than one per atom, and there are two types of carriers. Let the density of each carrier be n . The total carrier density is $2n$. Thus

$$\begin{aligned} \mu &= \sigma/2ne, \\ n\mu &= 1 \times 10^{27} \text{ (m V sec)}^{-1}. \end{aligned} \quad (11)$$

¹⁸ R. G. Chambers, in *The Fermi Surface*, edited by W. A. Harrison and M. B. Webb (John Wiley & Sons, Inc., New York, 1960), p. 100.

¹⁹ E. Fawcett and W. A. Reed, *Phys. Rev.* **134**, A723 (1964).

We now have two equations for the mobilities

$$\begin{aligned} \mu_e + \mu_h &= (10^{27}/n) \text{ m}^2/\text{V sec}, \\ \mu_e\mu_h &= 46 \text{ (m}^2/\text{V sec)}^2. \end{aligned} \quad (12)$$

Solving for the mobilities in terms of the carrier density n , we obtain

$$\mu_e, \mu_h = \frac{1}{2} \left[\frac{10^{27}}{n} \pm \left(\frac{10^{54}}{n^2} - 184 \right)^{1/2} \right]; \quad (13)$$

μ_e will take one sign on the radical, μ_h will take the other. Because the carrier density n appears in this equation, we cannot solve for the μ 's; we can, however, see that if n is larger than $7.4 \times 10^{19} \text{ cm}^{-3}$ then the discriminant will be negative. Thus an upper limit on the density of each carrier is $7.4 \times 10^{19} \text{ cm}^{-3}$. This would correspond to approximately 0.02 electrons per lithium atom. In the absence of good density data and sufficient knowledge of the crystal structure, further analysis is impossible.

It is unlikely that the high magnetoresistance is attributable to open orbits,¹⁸ since this scheme predicts a linear magnetic field dependence for the transverse magnetoresistance. Bowers²⁰ has recently reported a nonsaturating magnetoresistance in potassium. The field dependence is linear and the size of the effect is three orders of magnitude smaller than the present result.

SUMMARY

The electrical conductivity of $\text{Li}(\text{NH}_3)_4$ was measured from 2.9 to 200°K using an electrodeless technique. The material is bronze in appearance and metallic in behavior. The lithium tetrammine has a residual resistivity of $0.30 \mu\Omega\text{cm}$.

The magnetoresistance of $\text{Li}(\text{NH}_3)_4$ was measured at 4.2°K in magnetic fields up to 43 kG. It exhibits a large magnetoresistance never observed before in materials other than extremely pure metals. The coefficient of magnetoresistance is 0.46 (kG)^{-2} . The magnetoresistance is quadratically dependent on the magnetic field in the highest field available; it shows neither oscillatory behavior nor saturation. On the basis of the two-band model for magnetoresistance, $\text{Li}(\text{NH}_3)_4$ may be interpreted as a completely compensated metal such as Mo or W with equal electron and hole populations and closed Fermi surfaces for both the electrons and holes. Further work is in progress.

ACKNOWLEDGMENTS

The authors acknowledge with pleasure informative discussions with J. D. Gavenda and P. R. Antoniewicz. Data could not have been acquired in the 15–35°K range without the skilled and willing assistance of W. B. Wollet. Further technical assistance was provided by D. E. Bowen and R. B. Bell, among others.

²⁰ P. A. Penz and R. Bowers, *Bull. Am. Phys. Soc.* **11**, 92 (1966).