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Electron Spin Resonance in Metals and Metal-Ammonia Solutions

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Electron spin resonance measurements have been made at 300 Mc/sec on frozen metal ammonia solutions, liquid metal ammonia solutions, and liquid metals. The experiments on frozen solutions have shown that the metal is precipitated out in small particles. Nuclear resonance experiments have corroborated this finding. The frozen solutions are used as a convenient source of small particles, and observations have been made of relaxation times in Na, Li, K, and Cs. Resonances in the liquid ammonia solutions of Li, Na, K, Rb, Cs, and Ca, and also in a solution of Li in methylamine were observed. The data are consistent with the theoretical model of Kraus, Ogg, and Kaplan and Kittel. Bulk metal resonances were observed to temperatures above the melting points on lithium and sodium. The results for sodium corroborate the Elliott theory of relaxation, and the lithium results indicate resistivity-dependent relaxation time. No resonance was observed in bulk K, Rb, and Cs carried to temperatures above their melting points.

I. METAL-AMMONIA SOLUTIONS

A. Introduction

THAT ammonia dissolves sodium and potassium was first discovered by Weyl.[1] Subsequent work of Franklin, Kraus, and Walden and others established that all the alkalis and most of the alkaline earths were soluble in ammonia either by direct dissolution or electrochemically by discharge at the cathode.[2,3] The stability of the solutions decreases with increasing atomic weight, with a reaction of the form

$$X+\mathrm{NH_3}\rightarrow X\mathrm{NH_2}+\tfrac{1}{2}\mathrm{H_2}$$

taking place. Deterioration can be slowed by extreme purity of the ammonia and metals used to form the solution. Dewald and Lepoutre[4] have reported the lowest observable decomposition rate to be 0.1% per day. A model which describes the spin resonance and other properties has been discussed by Kaplan and Kittel.[2,5] The alkali atom upon dissolving becomes ionized, and the freed valence electron is then trapped in a vacancy of radius 4 A in the liquid. The volume of this hole is set equal to the observed volume increase on solution.

Extensive work has been done by Hutchison and Pastor[6] on electron spin resonance in solutions of sodium and potassium. They have investigated line width, intensity, g-values, and saturation in these solutions. They find a g-value of 2.0012 for potassium solutions and line widths as narrow as 0.02 oersted in dilute samples at 298°K.

The spin resonance line breadth is explained as follows. The electrons are assumed to occupy atomic orbital states on the protons surrounding the vacancy. Two sources of broadening exist: dipolar interactions among the paramagnetic electrons, and hyperfine interactions between the electrons and the protons of which they are the s-orbitals. At low concentrations the hyperfine interaction is dominant; it produces a distribution of lines whose envelope is observable. The width of the envelope is independent of concentration. It turns out to be about 10 oersteds. However, the rotation of the ammonia molecules in the liquid causes the electron to "see" its proton for a time small compared to the duration of its spin state. The reduction of the effective field due to motional narrowing results in a width of about 0.05 oersted, in agreement with experimental data. At higher concentrations the dipolar width begins to take over, but it also is motionally narrowed, in this case by the random diffusive motion of the liquid.

B. Preparation of Samples and Experimental Setup

All measurements were made at a frequency of 300 Mc/sec on resonance equipment using a transmission coaxial cavity operating in a *TEM* mode. Details of the apparatus have been previously described[7] in the literature.

The coaxial cavity required the use of special Pyrex containers. A schematic diagram is shown in Fig. 1(a). Sample holders had a usable volume of from 1 to 2 cc, and quantities of metal varied from 1 to 50 milligrams depending on concentration desired and atomic density of the metal solute.

A standard process was used in the preparation of metal-ammonia solutions. Tank ammonia was condensed with dry ice and acetone into a flask containing chunks of sodium metal. The sodium reacts with water, oxygen, and the ammonia itself until all impurities are reduced. Then the sodium dissolves in the now clean

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* Now at Texas Instruments, Inc., Dallas, Texas.

[1] W. Weyl, Ann. Physik **121**, 601 (1864).

[2] C. A. Kraus, *The Properties of Electrically Conducting Systems* (Chemical Catalog Company, New York, 1922).

[3] W. C. Johnson and A. W. Meyer, J. Am. Chem. Soc. **54**, 3621 (1932); McElroy, Kleinberg, and Davidson, J. Am. Chem. Soc. **72**, 5178 (1950).

[4] J. F. Dewald and G. Lepoutre, J. Am. Chem. Soc. **76**, 3366 (1954).

[5] J. Kaplan and C. Kittel, J. Chem. Phys. **21**, 1429 (1953). See also R. A. Ogg, J. Chem. Phys. **14**, 114, 295 (1946).

[6] C. A. Hutchison and R. C. Pastor, J. Chem. Phys. **21**, 1959 (1953).

[7] G. Feher and A. F. Kip, Phys. Rev. **98**, 337 (1955); G. Feher, Ph.D. thesis, University of California, 1954 (unpublished).

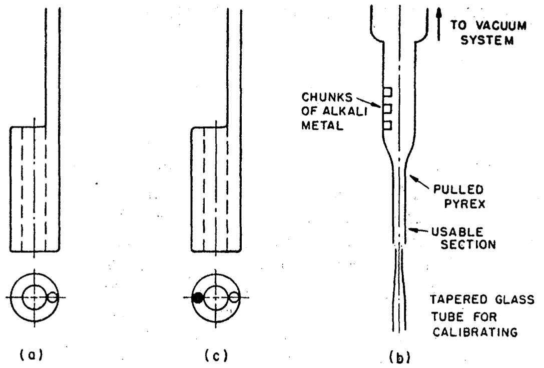

FIG. 1. Sample holders.

ammonia forming a deep blue solution. The existence of a blue solution is an excellent test of clean ammonia. Evolved gases are then removed by pumping the ammonia until it freezes. The ammonia remaining after pumping is condensed into calibrated sample holders containing the metal to be investigated.

Preparation of milligram quantities of alkali metal has been carried out as follows. Small-diameter glass capillaries are produced by pulling Pyrex tubing. See Fig. 1(b). The inside diameters are measured by the insertion of smaller tapered tubes. Chunks of alkali metal are introduced into the large end; the tube is evacuated, and the metal is melted. Helium gas is then used to force the liquid metal into the narrow section of the tube. The diameter can be made small enough so that 3-mg/cm sticks are obtained. To get a given weight of metal it is then necessary only to cut off a measured length.

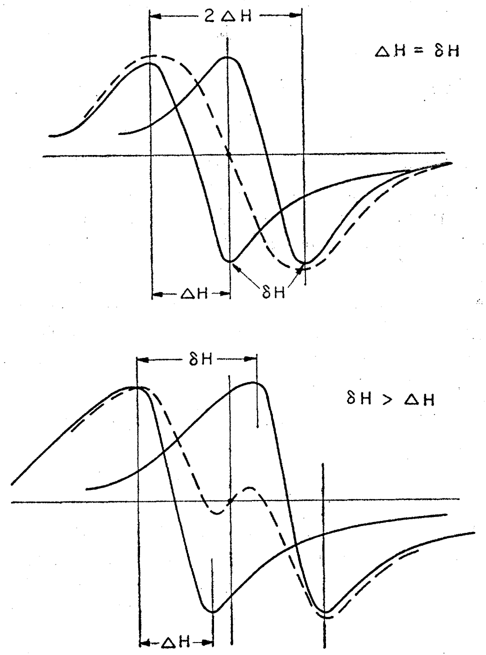

FIG. 2. Resolution of closely spaced derivative lines.

Since all the alkali metals were found to be inert at liquid nitrogen temperature, the breaking can be carried out easily in liquid nitrogen. This is much easier than using a dry box, and it is suggested as an easy way to handle alkali metal for any purpose. Even cesium metal shows no sign of reaction in liquid nitrogen.

After introduction of the metal into the sample holders, the containers are evacuated and then opened to condensation of ammonia. When the proper amount of ammonia has been condensed, the samples are sealed off and stored in liquid nitrogen to prevent the slow deterioration of the solutions which occurs, even at high purities,[4] since the solutions are thermodynamically unstable. Storage in the frozen state prevents any observable decomposition, and samples have been kept this way for several months.

C. Experimental Results

In addition to confirming Hutchison's and Pastor's observations of resonance in sodium and potassium, we have observed resonances in lithium, rubidium, cesium, and calcium solutions. Our data are shown in Table I.

TABLE I. Data in liquid metal-ammonia solutions.

Solution	ΔH peak-to-peak oersteds	Temperature °C	Concentration M
Li-NH_3	0.1	−72	0.2
Na-NH_3	0.13	−75	0.1
K-NH_3	0.1	−70	0.08
Rb-NH_3	0.16	−70	<0.1
Cs-NH_3	0.4	−70	0.5
Ca-NH_3	0.14	−70	0.05

We have compared directly g-values of the alkali and alkaline earth solutions. Samples of calcium in ammonia and sodium in ammonia were prepared and placed adjacent to each other in the cavity. A g-shift would have manifested itself as a splitting of the observed resonance line. No splitting was observed. Subsequent analysis of the resolvability of lines[8] (see Fig. 2) indicates that the difference in g-values between the sodium and calcium solutions is less than 0.005.

According to Kaplan and Kittel's theory of line width, the critical parameter is the frequency of the perturbation causing the width. In dilute solutions (below 0.1 M) the main perturbation is the hyperfine interaction, and the fluctuation frequency is given by the frequency of rotational dipolar relaxation. From

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[8] We consider two curves of equal amplitude and width (Fig. 2). As the sketch shows, if the spacing δH of two curves is greater than the peak-to-peak line width ΔH, they are resolvable. If $\Delta H = \delta H$, the envelope is unresolved but its width is $2\Delta H$. If $\delta H < \Delta H$, the envelope is still unresolved, but its width is given by $(\Delta H)_{\text{envelope}} = \Delta H + \delta H$. Therefore, if the individual widths are known, the spacing is calculable from the above equation.

Eq. (13) of reference 5, we have

$$\omega_c \approx kT/3\eta V, \qquad \text{(I.1)}$$

where η is the viscosity of the liquid and V is the molecular volume. The line width is inversely proportional to ω_c and therefore directly proportional to viscosity. Since the viscosity of liquids varies with temperature, the line width should vary accordingly. This is found to be the case. Figure 3 is a comparison of line width and viscosity as a function of temperature. They are normalized at -33.5°C, the boiling point of ammonia. The data plotted are from Fig. 5 of reference 6, using the 0.1 M concentrations and our data for potassium at 0.08 M.

A further test of theory presents itself in experiments on metal solutions of methylamine (CH_3NH_2). A sample of lithium of about 0.1 M concentration in methylamine was prepared by the same process used for the ammonia solutions. Measurements of spin resonance line widths were made at -80°C, -40°C, and 0°C, using dry ice and acetone, dry ice and dirty ethanol, and ice, respectively. Data for viscosity of methylamine are meager, only one point being known. $\eta = 0.236$ centipoise at 0°C. In order to obtain viscosity as a function of temperature from universal viscosity charts,[9] values of viscosity must be known at two temperatures. We have obtained the value of viscosity at -54°C from Shell Development Company. (We wish to thank Mr. G. C. Pounds of Shell Development Company for kindly making this measurement.) Figure 4 shows a comparison of the spin resonance line width and viscosity of methylamine as a function of temperature. The data are normalized at -80°C. At high temperatures the line width appears to be leveling out faster than the viscosity. This may be due to the dipolar contribution beginning to make itself felt.

It is of interest to compare the methylamine data with that of the ammonia to see whether the consistency persists. We compare results at -70°C. Experimentally we have

$$\Delta H_{\mathrm{NH_3}} \approx 0.1 \text{ oersted},$$

$$\Delta H_{\mathrm{CH_3NH_2}} \approx 0.6 \text{ oersted},$$

so

$$\Delta H_{\mathrm{CH_3NH_2}}/\Delta H_{\mathrm{NH_3}} \approx 6.$$

This is to be compared with the ratio of ηV for the two materials:

$$(\eta V)_{\mathrm{CH_3NH_2}}/(\eta V)_{\mathrm{NH_3}} = (0.7)(67)/(0.5)(34) \approx 3.$$

The agreement is fairly good since the implied assumption of the equality of the intrinsic widths (without motional narrowing) is probably not accurate. The larger size and ellipsoidal shape of the methylamine molecules might reduce the number of protons surrounding the e_1 center and produce a wider line. The ammonia molecule also possesses an extra degree of freedom (oscillation of the N ion in and out of the triangle of protons) which is not found in methylamine. This additional oscillation will increase the narrowing of the ammonia lines with respect to the methylamine.

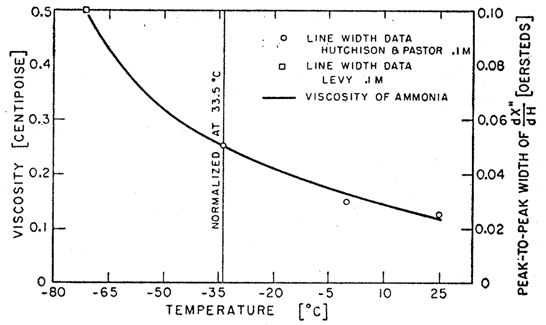

FIG. 3. Comparison of line width and viscosity as functions of temperature: liquid metal-ammonia solutions.

II. ALKALI METALS

In metals it is the spin-lattice relaxation time T_1 which determines the line width. Several mechanisms for spin-lattice relaxation have been proposed. Overhauser[10] has discussed several interactions of which the one giving rise to the shortest relaxation time is the interaction between the magnetic moment of one electron and the field caused by the translational motion of a second electron. Elliott[11] has discussed a process whereby the small amount of orbital angular momentum associated with the spin state causes the spin to flip. He obtains the following expression for the spin-lattice relaxation time:

$$\begin{aligned} T_1 &= \alpha \tau_R/(\Delta g)^2 \\ &\propto 1/[(\Delta g)^2 \rho]. \end{aligned} \qquad \text{(II.1)}$$

Therefore,

$$\Delta H \propto (\Delta g)^2 \rho.$$

The spin-orbit coupling enters through the g-shift Δg, which is proportional to λ/Δ, where λ is the spin-orbit coupling constant and Δ is the energy level separation.

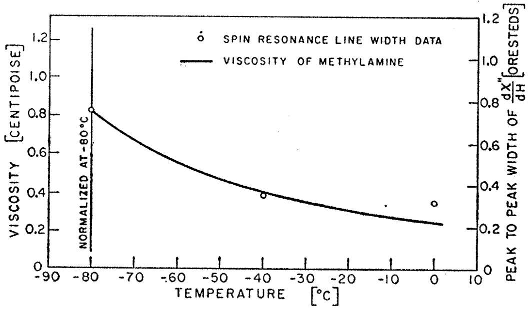

FIG. 4. Comparison of line width and viscosity as functions of temperature: liquid lithium-methylamine solution.

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[9] W. H. McAdams, *Heat Transmission* (McGraw-Hill Book Company, Inc., New York, 1954), third edition, p. 466.

[10] A. W. Overhauser, Phys. Rev. **89**, 689 (1953).

[11] R. J. Elliott, Phys. Rev. **96**, 266, 280 (1954).

The g-shift has been calculated for conduction electrons in several metals by Yafet[12] and Brooks.[13] τ_R is the mean time between collisions for electrons as determined by resistivity measurements; ρ is the resistivity in ohm-centimeters. α is a constant, of the order of 0.03 for phonon collisions. Yafet[14] has examined Elliott's assumptions, and he finds that the factor α is a function of temperature such that the relaxation has a $1/T$ dependence at low temperatures instead of $1/T^3$ as predicted by Elliott.

Experimental Procedure

Samples of bulk sodium and potassium were prepared by forcing molten metal into glass containers and subsequently sealing them. Samples of rubidium and cesium were obtained by sending our containers to A. D. MacKay Company to be filled. Figure 1(c) is a schematic diagram of the container.

Because of its reactivity with glass at high temperatures, lithium had to have special handling. After many unsuccessful attempts to force molten lithium into glass, the following procedure was devised. The glass container was first lined with Standard SAE 40 motor oil which had previously been heated at 220°C to remove volatile vapors. Then molten lithium was forced in under helium pressure and the container subsequently sealed.

Temperature ranges between 4°K and 500°K were studied. To produce these temperatures the following procedures were used:

1. 4°K to 77°K.—Liquid helium was introduced into the inner Dewar of a double Dewar system. The inner Dewar contained the cavity, so the cavity was in contact with liquid helium. The outer Dewar was filled with liquid nitrogen at 77°K. Measurements at 4°K were made with the cavity completely immersed in liquid helium. The range from 4°K to 77°K could be covered satisfactorily by taking data after the helium level fell below the bottom of the cavity and the cavity began to heat up. In the lithium-ammonia runs, the range from 65°K to 77°K was critical, and this region was covered nicely by pumping liquid nitrogen placed in the inner Dewar.

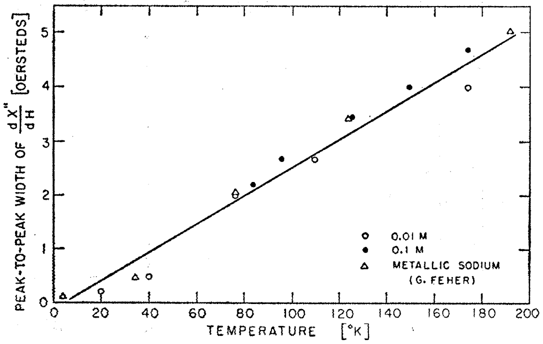

FIG. 5. Line width *versus* temperature: frozen sodium-ammonia solutions.

2. 77°K to 200°K.—This region was covered by having liquid nitrogen in the outer Dewar and heating the cavity electrically with a coil attached below its lower end. With practice we could obtain any desired ambient temperature in this region.

3. 200°K to 273°K.—This region was not covered continuously, but points at 200°K, 230°K, and 273°K were obtained by using dry ice and acetone, dry ice and watered ethanol, and an ice bath, respectively, in the outer Dewar. The inner Dewar in these runs was a single-walled Pyrex tube used only to keep the cooling bath out of the cavity.

4. 300°K to 500°K.—The single-walled inner Dewar was wound with a heating coil and placed in the outer Dewar filled with an oil having a high flash point (Standard, Pure Pennsylvania, SAE 40). Fifty watts was ample power to produce 500°K. Teflon insulation was used in the cavity coupling pieces.

Results in Frozen Metal-Ammonia Solutions

We find that metal-ammonia solutions, when frozen, become separated into metal particles and ammonia. Observations carried out on several alkali-ammonia solutions support this conclusion as shown below.

1. Sodium.—We observed sodium-ammonia solutions of 0.1 M and 0.01 M concentrations. Temperature dependence of the spin resonance lines was studied from 77°K to 220°K and from 4°K to 200°K, respectively. The 0.01 M sample cracked and decomposed when taken above the melting point. It may be mentioned that most samples taken to liquid helium were cracked after removal from the cavity. Presumably the fast cooling from 77°K to 4°K which accompanies a helium transfer caused the breakage. Therefore measurements in liquid solutions had to be made before any helium runs on the same sample. Figure 5 shows the data for line width between maximum slopes (hereafter referred to as peak-to-peak width) as a function of temperature and the same data for metallic sodium. The width of the absorption is independent of concentration and its

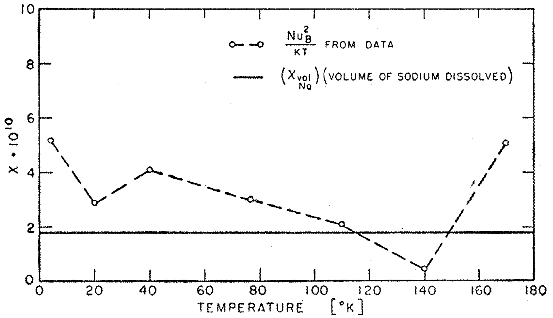

FIG. 6. Susceptibility of frozen sodium-ammonia solution.

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[12] Y. Yafet, Phys. Rev. **85**, 478 (1952).
[13] H. Brooks, Phys. Rev. **94**, 1411(A) (1954).
[14] Y. Yafet (unpublished).

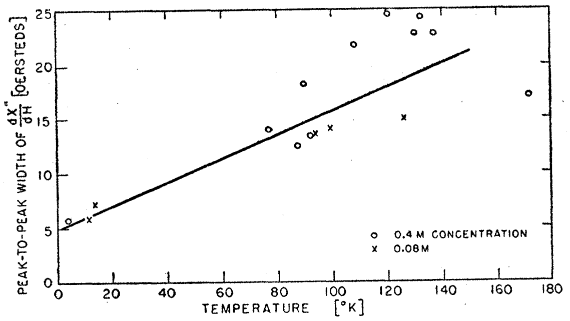

FIG. 7. Line width *versus* temperature: frozen potassium-ammonia solutions.

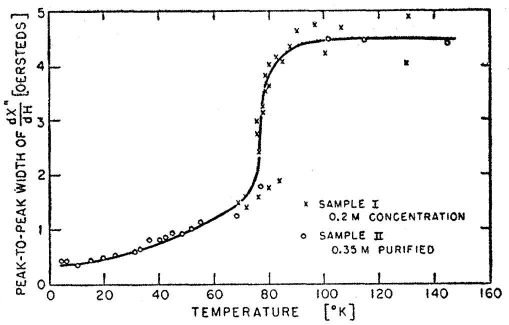

FIG. 8. Line width *versus* temperature: frozen lithium-ammonia solutions.

behavior coincides with that of metallic sodium. All lines observed were symmetrical absorptions having approximately Lorentzian shapes. These results agree with Yafet's prediction of a linear temperature dependence of line width.

The presence of a calibrated sample of diphenylpicrylhydrazyl in a cavity allowed a calculation of susceptibility as a function of temperature to be made. The standard sample was located so it did not overlap the resonance of the metal-ammonia sample. Results of the susceptibility determinations are shown in Fig. 6. Within the accuracy of the measurements the susceptibility is independent of temperature.

The nuclear resonance of frozen sodium-ammonia solution provided additional information. In 1949 Knight[15] observed that the nuclear resonance of several metals occurred at different frequencies (in the same magnetic field) than salts of the same isotope. This Knight shift has since been the subject of considerable experimental and theoretical investigation. It has been established that the shift is due to an effective field at the nuclei due to their hyperfine interaction with the conduction electrons of the metal, and therefore is able to give information about the electron wave functions in metals.

The discovery that the electron spin resonance in frozen metal ammonia solutions might be due to precipitated metal particles suggested that the appearance of the Knight shift in the nuclear resonance would be a confirming fact.

Accordingly, a sample containing 1 cc of ammonia and 10^{20} atoms of sodium was prepared, and Knight examined its nuclear resonance. An absorption appeared which had the shape, width, and shift characteristic of metallic sodium. Comparison with a standard sample of sodium metal showed a frequency difference of only 0.25 kc/sec, which is in agreement to one part in 25. No unshifted sodium line appeared, and since the signal-to-noise ratio in the sample signal was about ten, not more than 10% of the sodium atoms in the sample could have been in the nonmetallic state.

Further evidence of the nature of frozen metal-ammonia mixtures comes from the work of Bosch,[16] who made optical absorption measurements on alternate layers of ammonia and alkali metals evaporated onto a quartz plate at various temperatures. Her experiments indicate that below 60°K the metal remains dissolved in ammonia; above 60°K, precipitated metallic particles are formed.

This result is significant to the work described here for the following reason. In all of our experiments on frozen metal-ammonia solutions, the material was prepared from the liquid and subsequently quenched in liquid nitrogen (77°K). The metal atoms thus had plenty of time to coagulate to form metal particles. (Our experiments at 4°K were on samples which had previously been stored for at least a day at 77°K.)

2. *Potassium.*—Spin resonance measurements were made on two concentrations of potassium in ammonia, 0.08 *M* and 0.4 *M*. The temperature range from 4°K to 180°K was studied as shown in Fig. 7. All lines showed pure absorption behavior characteristics of particles small compared to a skin depth except for the 0.4 *M* sample at 4°K, where distortion began to appear. This is further evidence that the resonance is due to metal particles since the reduced skin depth in the metal at low temperature would cause this distortion.

The potassium line at 4°K is about 50 times as wide as sodium. This is somewhat smaller than the prediction on the basis of Elliott's equation (1.4), but the work of Yafet[14] indicates that the factor α can be different for different elements, so the experimental results are not inconsistent with theory.

Potassium-ammonia samples exhibited two characteristics which we cannot explain. The first is a decrease of the line width as the melting point is approached. This is seen in Fig. 8 by the points at 170°K. The other is an *increasing susceptibility* as temperature increases. That this was not a cavity effect or the effect of changes in instrument parameters was verified by comparison with a diphenylpicrylhydrazyl standard sample in the cavity.

3. *Lithium.*—Figure 8 shows the temperature dependence of line width in frozen lithium-ammonia

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[15] W. D. Knight, Phys. Rev. **76**, 1259 (1949).

[16] E. Bosch, Z. Physik **137**, 89 (1954).

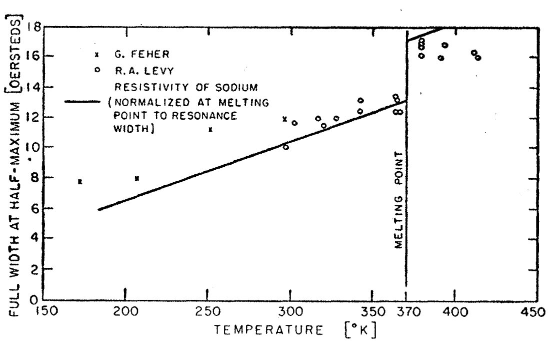

FIG. 9. Comparison of line width and resistivity as functions of temperature: bulk sodium metal.

solutions. The data presented are for two samples. Sample I is a 0.2 *M* concentration using 99.9% pure lithium obtained from Lithium Corporation of America. Sample II is a 0.35 *M* concentration using a distillate of the same material kindly provided by M. Browne.

Comparison of our data with results in metallic lithium are less fruitful than for sodium. The temperature dependence of line width in metallic lithium is anomalous with respect to the other alkalis and with respect to theories of relaxation in metals since it shows no temperature dependence. Lithium in bulk from the 99.9% batch generally shows a three-oersted line. The distillate shows a one-oersted line when evaporated out as a thin film. As the graph shows, both the bulk and the distillate, when dissolved in ammonia, show identical behavior.

The g-shift in lithium metal is calculated to be less than 10^{-4}, and in fact none has been observed.[7] On the basis of the calculated shift, the line width in lithium metal at 300°K is 0.05 oersted from the Elliott theory. Observed line widths have varied between 0.3 oersted and 10 oersteds depending on the purity and history[17] of the sample. The experiments of Feher and Browne support the hypothesis that impurity atoms such as sodium increase the spin-orbit interaction and in effect determine the line width.

As seen from Fig. 8, the line width in the frozen lithium-ammonia solutions remains constant above about 80°K just as in ordinary lithium metal. However, the abrupt transition to a narrower line has no counterpart in the bulk metal work. Barrett and Frank[18] have reported that a phase transition occurs in lithium at 75°K, the transition being from body-centered cubic to face-centered cubic as the temperature is lowered, and the transition of line width is probably connected with this change in structure.

It may be mentioned that the 0.35 *M* concentration showed the effect of particles large compared to a skin depth. All lines observed at this concentration had the $\chi'+\chi''$ shape.

4. Cesium.—A sample of 0.5 *M* cesium in ammonia was observed in the temperature region between 4°K and 77°K. A line 13 oersteds wide at $g=1.93\pm0.02$ was seen between 4°K and 25°K. (See Table II.) Above 25°K it could not be observed. Comparison with a standard hydrazyl sample indicated that the signal was not due to a paramagnetic impurity since the intensity did not follow a $1/T$ dependence but appeared constant throughout the observable range. Brooks' calculated g-value for cesium is 1.94, which seems to be in excellent agreement with the observations. There are no other data in metallic cesium with which to compare our results. We have also examined a sample of bulk cesium sealed in glass, but no resonance was observed in the temperature range between 4°K and 300°K.

Results in Metals at Elevated Temperatures

The experiments of Feher and Kip[7] provided the following information about spin-lattice relaxation times in metals:

1. Sodium.—The relaxation time is approximately proportional to conductivity from 4°K to 300°K.

2. Lithium.—The relaxation time is independent of temperature between 4°K and 300°K, but is longer the more highly purified are the samples.

The results in sodium appear to confirm the Elliott-Yafet mechanism of relaxation inasmuch as the line width follows the same law as does the resistivity. The data for lithium are a bit confusing since the highest purities obtained should have yielded intrinsic relaxation times.

Extension of the measurements to higher temperatures would provide a better check on the theory especially if the melting points of the metals could be exceeded, since there the resistivities have discontinuities. Measurements were carried out on all the alkali metals to temperatures above their melting points. Resonance was observed only in sodium and lithium as expected from the theory, since the spin-orbit interactions are too large in the other alkalis to produce observably narrow lines in the temperature ranges studied.

Figure 9 shows the dependence of line width and resistivity on temperature for sodium. The discontinuity in line width at the melting point provides a clear confirmation of the effect of resistivity on relaxa-

TABLE II. g-Values in frozen metal-ammonia solutions.

	Lithium	Sodium	Potassium	Cesium
Calculated	2.0023[a]	2.002[b]	1.998[c]	1.94[c]
Observed	2.00	2.00	1.99	1.93

[a] P. N. Argyres and A. H. Kahn, Phys. Rev. 98, 226(A) (1955).
[b] See reference 12.
[c] See reference 13.

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[17] M. Browne (unpublished work).

[18] C. S. Barrett and O. R. Frank, Trans. Am. Inst. Mining Met. Engrs. 175, 579 (1948).

tion time. All the lines had the characteristic shape exhibited by metal samples large compared to a skin depth.[19] At these temperatures the effects of spin diffusion[7,20] are unimportant. The intensity of signals remained constant throughout the temperature range examined. Failure to observe a decrease in intensity above the melting point due to the increased line breadth may be accounted for by the larger volume of metal presented to the rf field.

The results in lithium appear in Fig. 10. The characteristic temperature-independent width is observed to persist to the melting point. The discontinuity above the melting point was reversible, that is the sample on cooling returned to its original width. This removes the possibility that the increased width was due merely to increasing diffusion of impurities into the sample from the container.

If we assume that the observed three-oersted line at 200°C is intrinsic, the calculated intrinsic width at room temperature from the resistivity change is 0.6 oersted. Feher has observed a temperature-independent 0.3-oersted line at room temperature and below in an evaporated film. We may tentatively conclude that the intrinsic width has been measured at room temperature. The factor of 2 discrepancy may be ascribed to Yafet's suggestion that at high temperatures imperfections may be more important to the relaxation than phonons, so line width may go up faster than resistivity.

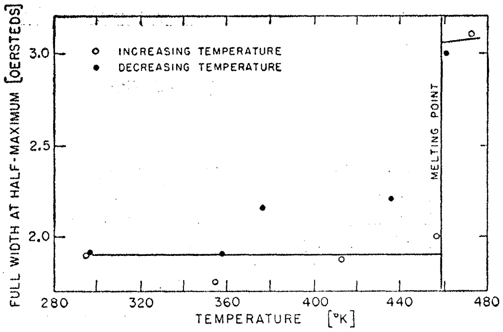

FIG. 10. Line width *versus* temperature above 300°K: bulk lithium metal.

Potassium, rubidium, and cesium were studied to temperatures exceeding their melting points, but no resonance was observed in any of the samples.

ACKNOWLEDGMENTS

It is a pleasure to express my sincere appreciation for the helpful guidance of Professor A. F. Kip and Professor C. Kittel under whose direction this work was performed. I would like to thank Dr. G. Feher for many pleasant, profitable, and instructive conversations. I am indebted to Professor W. D. Knight who kindly made nuclear resonance measurements, to L. Griffith who generously made available for my use his liquid ammonia system, and to Professor R. A. Ogg for instructive conversations on metal-ammonia techniques. I wish to express my gratitude for the assistance and support of the Office of Naval Research, the U. S. Signal Corps, and the Pittsburgh Plate Glass Foundation.

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[19] N. Bloembergen, J. Appl. Phys. 23, 1383 (1952).

[20] F. J. Dyson, Phys. Rev. 98, 349 (1955).