

berkelium isotopes in a windowless gas proportional counter showed the 4.6-hour Bk<sup>243</sup> previously reported<sup>3</sup> and a considerably longer-lived decay period corresponding to a  $4.95 \pm 0.1$ -day half-life. Some of this same radioactivity was examined in a differential alpha-pulse analyzer,<sup>4</sup> which revealed alpha-particles of the following energies and abundances:  $6.33 \pm 0.05$  Mev (18 percent),  $6.15 \pm 0.05$  Mev (48 percent), and  $5.90 \pm 0.05$  Mev (34 percent). All three alpha-particle groups were observed to decay with a 4.95-day half-life. Initially the three alpha-particle groups belonging to the 4.6-hour Bk<sup>243</sup> were also present. Consideration of the systematics of alpha-radioactivity<sup>5</sup> suggests that the new 4.95-day isotope is most likely Bk<sup>245</sup>. The new berkelium isotope must be of mass greater than 244 or it would have been observed in bombardments of Am<sup>241</sup> with alpha-particles. Furthermore, much longer half-lives than 4.95 days were predicted for Bk<sup>246</sup> and Bk<sup>247</sup> using methods of estimation which have been described previously.<sup>6</sup> Comparison of the counting rate of the 4.95-day radioactivity in the windowless counter to that in the alpha-pulse analyzer allows calculation of  $\sim 0.1$  percent alpha-branching, which corresponds to a 15-year partial alpha-half-life. The alpha-particle decay of this isotope appears to be hindered by a factor of  $\sim 100$  (using the conventions of reference 5) and resembles the decay of the isotope Bk<sup>243</sup> in this respect.

Following the decay of the berkelium isotopes, the curium daughters resulting from electron capture decay were examined in the alpha-pulse analyzer and the characteristic alpha-particles of Cm<sup>243</sup> produced by the decay of 4.6-hour Bk<sup>243</sup> were observed. The amount of alpha-activity of energies expected for Cm<sup>245</sup>, namely, that in the range 5.4–5.8 Mev, was such that the half-life of the latter isotope must be at least 500 years if the energy is as indicated by the alpha-decay systematics.<sup>5</sup> This tentative conclusion is, of course, also based on the assumption that the mass assignment of the 4.95-day berkelium isotope is correct.

Examination of the californium fraction in the alpha-pulse analyzer soon after irradiation showed only 6.75-Mev alpha-particles decaying with a 35-hour half-life (tentatively assigned previously to Cf<sup>246</sup>).<sup>7</sup> The growth of alpha-particles of 6.08-Mev energy was observed and the amount corresponded to a half-life of approximately 160 days. This daughter is undoubtedly Cm<sup>242</sup> produced by the decay of Cf<sup>246</sup>, and the assignment to Cf<sup>246</sup> is now regarded as certain. The decay of the Cf<sup>246</sup>, followed through a decay factor of approximately 50, gave a half-life of  $35.7 \pm 0.5$  hours in agreement with previous measurements. The ratio of the counting rate in the windowless proportional counter to the alpha-counting rate corresponded closely to that observed for a number of "pure" alpha-emitters previously examined under similar conditions, so that Cf<sup>246</sup> appears to be beta-stable as expected. No radioactivity other than Cf<sup>246</sup> and its daughter Cm<sup>242</sup> was observed in the californium fraction. This result is somewhat surprising since it might have been expected that Cf<sup>247</sup> and possibly Cf<sup>245</sup> would have half-lives long enough to allow their observation.

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## The Magnetic Moment of As<sup>75</sup>\*

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NUCLEAR magnetic resonances of As<sup>75</sup> with a natural line width at half-maxima of about eight gauss have been observed in a solution of 2 molar Na<sub>2</sub>HAsO<sub>4</sub> in 3 molar NaOH in which the compound Na<sub>3</sub>AsO<sub>4</sub> is presumably formed. The resonant frequency was compared to that of Na<sup>23</sup> present in the above solution with the result

$$\nu(\text{As}^{75})/\nu(\text{Na}^{23}) = 0.64745 \pm 0.00015. \quad (1)$$

Using the fact that the spin of As<sup>75</sup> is  $\frac{3}{2}$ ,<sup>1</sup> and calculating the moment of Na<sup>23</sup> by taking  $\mu(H') = 2.79268 \pm 0.00006$  nuclear magnetons,<sup>2</sup> and  $\nu(\text{Na}^{23})/\nu(H') = 0.26450 \pm 0.00003$ ,<sup>3</sup> the sign and value of the magnetic moment was found to be

$$\mu(\text{As}^{75}) = +1.4347 \pm 0.0003. \quad (2)$$

The earlier determination of  $\mu(\text{As}^{75}) = 1.5 \pm 0.3$  nm by Schüler and Marketu<sup>4</sup> is in agreement with the more precise value of Eq. (2).

In order to avoid excessive line broadening due to the large quadrupole moment ( $+0.3 \times 10^{-24}$  cm<sup>2</sup>)<sup>4</sup> of arsenic, it is necessary to place the nucleus in a symmetrical molecular configuration. The solutions of the above proportions were chosen on the assumption that AsO<sub>4</sub><sup>-3</sup> ions of tetrahedral symmetry are formed.

Arsenic signals were detected in both liquid and solid phases of the above sample with a shift of about  $1/10^4$  towards higher fields for the solid state. Nuclear resonances of As<sup>75</sup> were not observed in other arsenic compounds (e.g., Na<sub>2</sub>HAsO<sub>4</sub>). This was probably due to the fact that the line widths, resulting from quadrupole effects, were too large.

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## The Electrical Conductivity of Liquid Germanium\*

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OBSERVATIONS of the melting of germanium have shown that the fusion process for this metal is very unusual. The entropy of fusion is 5.9 cal/deg,<sup>1,2</sup> about three times the value for most metals, and there is a considerable volume contraction on melting.<sup>3</sup> Solid germanium crystallizes in the diamond lattice, while the x-ray experiments of Hendus<sup>4</sup> show that the atomic arrangement in liquid germanium corresponds to a more nearly close-packed structure. These facts indicate that the binding forces of liquid germanium are of more metallic character than those of the solid, and lend interest to a measurement of the electrical conductivity of the liquid.

The difficulty of finding electrodes suitable for use with germanium at the high temperature required for this experiment necessitated measuring the effect of currents induced in a germanium core on the impedance of a surrounding coil. Unfortunately, the effects of temperature on the coil severely limit the accuracy of this technique.

The germanium was obtained from the Eagle-Picher Company and was stated to be of purity in excess of 99.9 percent. The sample container was made of quartz, and the experiment was performed in an argon atmosphere. Measurements were made at frequencies around 20 kilocycles and around 2 megacycles.

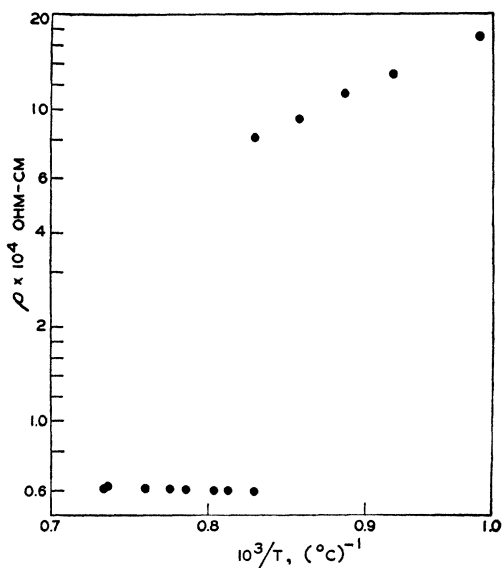


FIG. 1. Resistivity of germanium at high temperatures. The values for the resistivity of the solid are in good agreement with values obtained by extrapolating the direct current resistivity measured at lower temperatures.

The resistivity of the liquid germanium near the melting point was found to be  $60 \times 10^{-8}$  ohm-cm and the temperature coefficient of resistivity was found to be positive and about  $1$  or  $2 \times 10^{-4}$  (deg C) $^{-1}$ . A synthesis of data from several runs is shown in Fig. 1.

The melting of germanium thus shows a certain analogy to the transition from gray, semiconducting tin to white, metallic tin at  $13^{\circ}\text{C}$ . In the case of tin there is a 20 percent increase in density and an increase of the coordination number from four to six as the temperature is raised through the transition, and recent work<sup>5</sup> has shown that the resistivity decreases by a factor of 16. Estimates of the density of liquid germanium by the author based on Hendus' data give a density increase of about 20 percent in germanium upon melting, and the conductivity measurement shows that the resistivity of the solid germanium at the melting point is fifteen times that of the liquid.

The author is indebted to Dr. A. W. Lawson for encouragement and advice. Since the start of these experiments it has come to his attention that independent work on the same subject is being carried on by A. Epstein and Dr. K. Lark-Horovitz at Purdue University.

\* The investigations described were actually carried out in the laboratories of the Institute for the Study of Metals.

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### Local Fields in Ionic Crystals\*

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THE polarization of ionic crystals can consist of two parts. One part, the electronic polarization, arises from the polarization of the electronic distributions of the individual ions. The other part, the ionic polarization, arises from the displacement of the entire ions from their original positions in the lattice. When only electronic polarization is present, the local fields are found by replacing the electronic polarization with point dipoles at the lattice points. It has been customary in cases in which ionic polar-

ization is also present to adopt a similar procedure. In this procedure the electronic and ionic polarizations are replaced with point dipoles at the original lattice points and the local fields at the original lattice points are found. However, the use of the local fields at the original lattice points is inconsistent with ionic crystal theory and can lead to large errors. In ionic crystal theory the local fields at the nuclei of the ions must always be taken as the effective polarizing fields for both electronic and ionic polarization. Consequently the local fields at the displaced positions of the ions must be used when ionic polarization is present.

The difference between the local field at the original and that at the actual position of an ion may be of the same order of magnitude as either local field. To understand this qualitatively, consider a crystal  $AB$  in which the lattice  $A$  has undergone a small homogeneous displacement. The usual procedure for finding the local field for an  $A$  ion includes contributions at the original lattice point from "equivalent dipoles" on  $A$ . Actually, since  $A$  has only suffered a uniform translation, it will give no contribution to the local field at an  $A$  ion. The field on  $A$  results from the displacement of  $B$  relative to  $A$ . In calculating this field the equivalent dipoles should be put on  $B$  rather than on  $A$ . Thus, putting the dipoles on  $A$  may lead to significant errors in the local field.

For simplicity we omit explicit consideration of the contribution of the electronic polarization to the local field. The ionic contribution to the local field at the actual position of the  $i$ th ion is given by

$$\mathbf{E}_i = \sum_j' (z_j e \mathbf{r}_{ij}) r_{ij}^{-3}, \quad (1)$$

where  $z_j e$  is the charge of the  $j$ th ion and  $z_j$  its valence, and where the summation extends over all ions except the  $i$ th.  $\mathbf{E}_i$  vanishes for a crystal in which each lattice point is a center of charge, that is, in which there is no ionic polarization. Suppose that each sublattice of such a crystal experiences a small homogeneous displacement from its original position in the lattice relative to some one of the sublattices. The ionic polarization of the  $l$ th sublattice produced by its displacement  $\mathbf{d}_l$  is  $\mathbf{P}_l = n_l z_l e \mathbf{d}_l$ , where  $n_l$  is the number of ions per unit volume on the  $l$ th sublattice. The separation between ions is now  $\mathbf{r}_{ij} = \mathbf{r}_{ij} - (\mathbf{d}_j - \mathbf{d}_i)$  where  $\mathbf{r}_{ij}$  is their separation in the original lattice. The right side of (1) can now be expanded to first order in the displacements, yielding

$$\mathbf{E}_i = \sum_j' \{ 3z_j e [(\mathbf{d}_j - \mathbf{d}_i) \cdot \mathbf{r}_{ij}] r_{ij}^{-3} - z_j e (\mathbf{d}_j - \mathbf{d}_i) r_{ij}^{-5} \}. \quad (2)$$

The right side of (2) can be converted into a sum over sublattices:

$$\mathbf{E}_i = \sum_l' f_{kl} \mathbf{P}_l + g_k \mathbf{P}_k, \quad (3)$$

where

$$g_k = - \sum_l' f_{kl} (n_l z_l / n_k z_k). \quad (4)$$

Only those crystal structures will be considered for which the  $f_{kl}$  are simply constants, the dipole field constants of the original lattice. Equation (3) gives the local field at the actual position of the ion when ionic polarization is present. The first term in (3) is the contribution of the ionic polarization of all sublattices except the  $k$ th. It can be represented as due to equivalent dipoles at the original positions of these sublattices. The second term in (3) is the contribution of the ionic polarization of the  $k$ th sublattice itself. As shown by (4) this contribution can be interpreted as due to appropriate equivalent dipoles at the original lattice points of the other sublattices.

Taking the local field at the original position of the ion is equivalent to neglecting  $\mathbf{d}_i$  in (2). One would then have obtained  $\mathbf{E}_k = \sum_l' f_{kl} \mathbf{P}_l$ . The difference between the two local fields is  $(g_k - f_{kk}) \mathbf{P}_k$ . For sublattices for which  $g_k = f_{kk}$ , they are the same. The simplest crystals for which this is so are those with the NaCl structure and those with the CsCl structure. For many crystal structures, however,  $g_k - f_{kk}$  is of the same order of magnitude or greater than  $f_{kk}$  for one or more ions. The usual treatment of situations in which ionic polarization occurs must then be replaced by one in which the ionic polarization is dealt with explicitly. In particular, if only one sublattice has appreciable ionic polarization, a polarization equation for this ionic polarization must also be included in the treatment of the dielectric properties of the crystal. Calculations of this nature for  $\text{BaTiO}_3$  are discussed in the follow-