

cm<sup>2</sup>. A grain boundary was subsequently found stretched over the entire cross section of the wire at the base of one of the notches. This boundary, if free to move during the recrystallization process, would have bulged until  $p$  was equilibrated by the surface tension of the boundary. We estimated from Eq. (2), taking  $\alpha \sim 0.1$ , that  $p \sim 10^3$ . No bulging was detected. The sensitivity of the experiment to bulging of the boundary, however, when converted into pressure by assuming a grain-boundary energy of 500 dynes/cm, was also  $\sim 10^3$  dynes/cm<sup>2</sup>.

One may show directly from a momentum balance argument, which does not invoke the concept of  $\alpha$ , that  $p = n\epsilon l p_{GB}$ , a relation which is consistent with Eq. (1) and Eq. (2). Thus if the entire residual resistance in Blewitt's experiments were due to boundary scattering, and since there should be no significant difference between Cu and Al in this respect, we should have expected in our experiment an easily observable pressure of  $10^6$  dynes/cm<sup>2</sup>.

In conclusion, both the high residual resistance of Blewitt's recrystallized specimens and our negative experimental results indicate that some imperfections other than grain boundaries are present in Blewitt's recrystallized specimens. In view of the known sensitivity of resistance minima to impurities, and recalling Eq. (1), we suggest that an attempt be made to study the residual resistance as a function of grain size. Such a study, however, could not distinguish between pure grain-boundary effects and the effects of segregation of impurities at the boundaries.

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<sup>2</sup> J. S. Koehler, Phys. Rev. **94**, 1071 (1954).

<sup>3</sup> See, for example, J. Korrington and A. N. Gerritsen, Physica **19**, 457 (1953).

### Low-Temperature Thermal Conductivity of a Canadian Natural Graphite\*

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THE low-temperature thermal conductivity of artificial graphite and samples of small crystallite natural graphite have a temperature dependence<sup>1-3</sup> as

great as  $T^{2.8}$ . This is anomalous with regard to the  $T^2$  dependence of the specific heat.<sup>4</sup> Two interpretations of this anomaly have appeared. One interpretation attributes it to intergranular, nongraphitic carbon.<sup>3</sup> The other assumes it to be intrinsic to graphite but to be dependent on the ratio of crystallite width to thickness.<sup>5</sup> In order to distinguish between these interpretations the thermal conductivity of nearly single crystal graphite should be measured. A temperature dependence of  $T^2$  would be predicted by the first theory<sup>3</sup> and a higher exponent by the second.<sup>5</sup>

Since large crystals of graphite are not available, an apparatus was constructed to measure small flakes of natural graphite. The apparatus consists of a small carbon resistor heater, a heat meter (consisting of a sample of artificial graphite previously measured on another apparatus), and a flake (1/4 in.  $\times$  1/20 in.  $\times$  1/100 in.) of Canadian natural graphite<sup>6</sup> connected in series. The graphites were copper plated at the ends and soldered with Cerrolow - 117°F solder. Chromel-constantan thermocouples (0.001 inch diameter) were cemented onto the graphites to measure the temperature gradients. The whole is covered with a radiation shield and placed inside an evacuated vessel with temperature control between 4° and 300°K. The graphite samples used have deformation lines on the surface indicating crystallites of the order of  $10^{-2}$  cm. The electrical resistivity of these samples has a positive temperature coefficient similar to that of single-crystal graphite rather than artificial graphite.

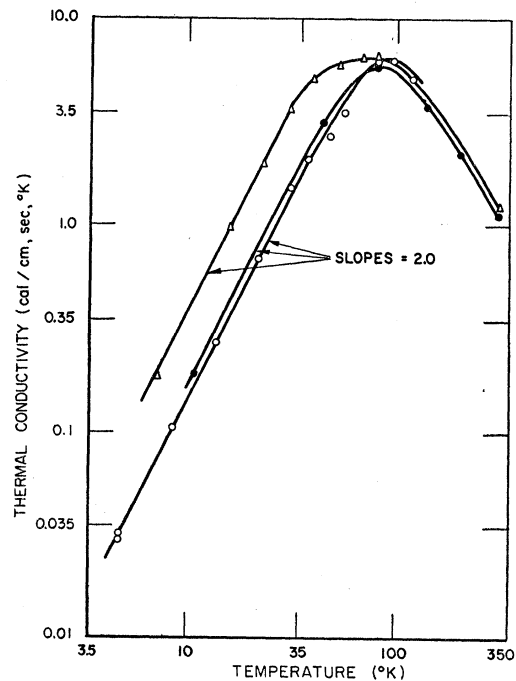


FIG. 1. Thermal conductivity of Canadian natural graphite.

The thermal conductivity of three samples is shown in Fig. 1. Due to the small size and irregular nature of the flakes, the absolute magnitudes are only approximate. The relative values are good to 5–10 percent. The maxima of the conductivity occur at around 80°K. Previously measured samples have maxima above room temperature. From this fact or from the actual conductivities at low temperatures, it is found that the mean free path, when boundary limited, is about 100 times that of any previous sample and slightly less than the crystallite size mentioned previously. The important fact, however, is that all three samples show a  $T^2$  dependence at low temperatures. This leads to the conclusion that the intrinsic conductivity does have a  $T^2$  dependence and substantiates the two-medium explanation of the higher exponent previously measured.

\* This report is based on studies conducted for the U. S. Atomic Energy Commission.

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<sup>2</sup> W. W. Tyler and A. C. Wilson, Jr., Phys. Rev. **89**, 870 (1953).

<sup>3</sup> A. W. Smith, Phys. Rev. **93**, 952 (1954).

<sup>4</sup> W. DeSorbo and W. W. Tyler, Phys. Rev. **21**, 1660 (1953).

<sup>5</sup> P. G. Klemens, Australian J. Phys. **6**, 405 (1953).

<sup>6</sup> Obtained from J. C. Bowman of the National Carbon Company.

### Paramagnetic Resonance Absorption in Sodium Plutonyl Acetate\*

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WE have observed paramagnetic resonance absorption in pure single crystals of  $\text{NaPuO}_2 \times (\text{C}_2\text{H}_3\text{O}_2)_3$  at 4°K and at a frequency of  $2.3 \times 10^{10} \text{ sec}^{-1}$ . The magnetic field was varied from 0 to  $1.4 \times 10^4$  gauss.

The Pu is contained in the four linear triatomic  $\text{PuO}_2^{++}$  molecules which are located on the body diagonals of the cubic unit cell of this compound.<sup>1</sup> Measurements have been made on three crystals. Each was rotated about an axis which was parallel to the  $\gamma f$  field. The three axes of rotation were the 112, 111, and 100 axes of the cube. Observations have been made of peak positions for 21 different orientations of the  $\text{PuO}_2^{++}$  axis ranging from  $\theta=0.108$  to  $\theta=0.425$ . A number of the angles were checked on more than one crystal. The positions are given very closely by  $g=5.92 \times |\cos\theta|$ , where  $g$  is  $\hbar/\beta$  times the ratio of frequency to field strength at maximum absorption and  $\theta$  is the angle between the  $\text{PuO}_2^{++}$  axis and the magnetic field. This corresponds to  $g_{11} \cong 5.9$ ,  $g_{12} \cong 0.0$ .

This behavior is that to be expected for an electronic structure of  $\text{PuO}_2^{++}$  in which the two electrons not in closed shells are  $f$  electrons.<sup>2</sup> In the strong axial electric field of  $\text{PuO}_2^{++}$ , the two electrostatically interacting  $f$  electrons will be in states with  $M=\pm 3$  and  $M=\pm 2$ . The spin, 1, will interact with the state  $M=5$  so that  $L_z+2S_z=\pm 3$  for the lowest doublet and hence the magnetic splitting will be  $6\beta H$  when  $H$  is parallel to the molecular axis and zero when it is perpendicular. Under these assumptions or taking the trigonal character of the field into account there would be no observable resonance absorptions because of the vanishing of the matrix elements of magnetic moment between the split states. Slight departures from the ideal symmetry apparently permit the observation of the transition.

The powder susceptibility would be given to first order at low temperatures by

$$\chi = \frac{N\beta^2}{kT} \left( \frac{g_{11}^2 \pm 2g_{12}^2}{12} \right) = \frac{1.09}{T},$$

corresponding to  $3\chi kT/N\beta^2=2.95$ . This is to be compared with the measured value 2.83<sup>3</sup> for  $\text{PuO}_2^{++}$  and 3.17<sup>4</sup> for the presumably isoelectronic  $\text{NpO}_2^+$ .

The resonance data offer convincing evidence that the two unpaired electrons in the plutonyl compound are  $f$  electrons.

A 2 percent solution of  $\text{NaPuO}_2(\text{C}_2\text{H}_3\text{O}_2)_3$  in the isomorphous diamagnetic  $\text{NaUO}_2(\text{C}_2\text{H}_3\text{O}_2)_3$  has been investigated and the nuclear hyperfine structure has been observed. In one orientation a doublet (the nuclear spin of Pu is  $\frac{1}{2}$ )<sup>5</sup> with a spacing of 430 gauss was found. The hyperfine structure data have not yet been interpreted.

We thank the Institute for the Study of Metals of the University of Chicago for supplying the liquid helium used in these experiments. We thank Mr. John Anderson for assistance in part of the experimental work. Mr. Edward Bartal constructed some of the apparatus employed in the measurements.

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<sup>2</sup> The theory of resonance in such ions is discussed in detail by J. Eisenstein and M. H. L. Pryce in a forthcoming paper. We wish to thank them for communicating their results to us in advance of publication. Various models of the situation resulting from crystalline field and spin-orbit perturbations of heavy metal ion electronic states have been discussed by R. J. Elliott, Phys. Rev. **89**, 659 (1953) and by D. M. Gruen and C. A. Hutchison, Jr., reference 4.

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<sup>5</sup> Van den Berg, Klinkenberg, and Regnaut, Physica **20**, 37 (1954).