

change in this temperature range.<sup>4</sup> Divalent metals, such as cadmium, may be expected to show a low density of states at the Fermi surface and effects due to the Brillouin zone boundaries, such as small pockets of low-effective-mass electrons and holes. The explanation of the temperature dependency will undoubtedly be found in the behavior of these electrons. At present one must consider both the possibility of an explicit temperature dependence of magnetic susceptibility and an indirect temperature dependence through changes in the crystal parameters.

The anisotropy of the temperature coefficient of expansion in cadmium is quite large, so that one might suspect a relation between the  $c/a$  ratio and the  $K$  values. That is, the effective mass discussed above might change with the  $c/a$  ratio due

to changes wrought in the Fermi surface at its intersection with the Brillouin zone boundary. An investigation has thus begun of the effects of decreasing the  $c/a$  ratio by adding magnesium.

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<sup>3</sup>J. Marcus, Phys. Rev. **76**, 621 (1949). The author would like to thank J. Butterworth for calling attention to this work.

<sup>4</sup>H. H. Landolt and R. Börnstein, Zahlenwerte und Funktionen aus Physik·Chemie·Astronomie·Geophysik und Technik (Springer-Verlag, Berlin, 1955), Vol. 2, Pt. 6, p. 35.

### LATTICE VIBRATIONS IN DIAMOND\*

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The purpose of this Letter is to report measurements of transverse acoustic (TA) dispersion curves for lattice vibrations propagating in the (100) and (111) directions in diamond. The measurements were made by the method of inelastic scattering of slow neutrons. The observed dispersion curves are in good agreement with those inferred from specific-heat and infrared absorption measurements but disagree with those deduced by assuming that diamond is homologous to silicon and germanium.

The experiment was carried out on a three-axis neutron diffraction spectrometer at the Los Alamos Omega West reactor. A version of the "constant  $Q$ " method was used in which the neutron's initial energy was fixed and its energy loss varied.<sup>1</sup> All measurements were carried out at room temperature. The sample was a 242.80-carat brown industrial diamond.<sup>2</sup> A neutron diffraction study showed it to consist of a large single crystal, with a mosaic spread of  $\approx 1^\circ$  full width at half-maximum, plus two small regions misoriented by  $3\text{--}5^\circ$ . The intensity of Bragg reflections from the two misoriented regions was less than 2% of that of corresponding reflections from the main body of the crystal. Infrared and ultraviolet absorption measurements indicated that it was a type-IIa diamond. The experimental data are given in Table I. Uncertainty in the observed frequencies, as estimated from the point-to-point

fluctuations and reproducibility of the data, is  $\approx \pm 0.2 \times 10^{13}$  rad/sec. We believe that possible systematic errors should at worst double this figure.

Except at a very high temperature, the specific heat of diamond is completely dominated by the critical point of the lowest dispersion curve, which is the zone boundary point of TA(111). Using this idea, Phillips has fit the specific heat of diamond with a model which gives a value  $\omega$

Table I. Frequencies (in units of  $10^{13}$  rad/sec) vs reduced wave number  $Z = q/q_{\max}$  for transverse acoustic lattice vibrations in diamond.  $q = 2\pi/\lambda$ . In the (100) direction  $q_{\max} = 1.7614 \times 10^8 \text{ cm}^{-1}$ ; in the (111) direction  $q_{\max} = 1.5254 \times 10^8 \text{ cm}^{-1}$ . The lattice constant of diamond was taken to be  $3.5672 \text{ \AA}$ .<sup>a</sup>

$Z$	$\omega_{\text{TA}(100)}$	$\omega_{\text{TA}(111)}$
0.3	6.45	5.38
0.4	8.63	6.76
0.5	10.24	7.96
0.6	11.53	8.80
0.7	12.74	9.51
0.8	13.70	9.99
0.9	14.59	10.26
1.0	15.13	10.38

<sup>a</sup>International Tables for X-Ray Crystallography, edited by J. S. Kasper and K. Lonsdale (The Kynoch Press, Birmingham, England, 1962), Vol. III, Chap. 2, p. 122.

$= 9.5 \times 10^{13}$  rad/sec for the zone-boundary frequency of the TA(111) curve.<sup>3</sup> This is in reasonable agreement with our measured value of  $10.4 \times 10^{13}$  rad/sec.

Hardy and Smith have measured the infrared absorption spectrum of diamond and from their results have constructed a set of dispersion curves for diamond which are in qualitative agreement with our results.<sup>4</sup> Bilz, Geick, and Renk have reviewed all the available data on lattice vibrations and infrared absorption in germanium, silicon, and diamond and have predicted that the zone-boundary frequency for the TA(100) curve should be  $15.1 \times 10^{13}$  rad/sec.<sup>5</sup> This is in exact agreement with our results.

Diamond, silicon, germanium, and gray tin are all homopolar crystals having the same structure. In each, the binding is attributed to the same tetrahedral arrangement of covalent bonds. It would seem reasonable to make the hypothesis that the interatomic restoring forces should also be the same. It would then follow that the difference in the observed dispersion curves should depend only on the difference in atomic mass and lattice spacing. On the basis of this homology argument and the observed dispersion curves for silicon<sup>6</sup> and germanium,<sup>7</sup> Kucher has postulated that all diamond-type crystals have the same dispersion curves when the frequencies are expressed in dimensionless units.<sup>8</sup>

Following Kucher, we have converted the observed frequencies for diamond, silicon, and germanium to dimensionless units  $\Omega = (Ma^3/2e^2)^{1/2}\omega$ , where  $M$  is the mass of an atom,  $d = 2a$  is the lattice space,  $e$  is the electronic charge, and  $\omega$  is the observed frequency. For silicon, we have used the recent measurements reported by Dolling.<sup>9</sup> In Fig. 1 we have plotted  $\Omega$  against the reduced wave number  $Z = q/q_{\max}$ . It may be easily seen that while the data for silicon and germanium are indeed very similar in this representation, those for diamond differ widely from the other two. Other scale factors have been used in comparing dispersion curves for these elements: Brockhouse<sup>6</sup> multiplies observed frequencies by  $(Ma^2)^{1/2}$ ; Bilz, Geick, and Renk divide by the Raman frequency<sup>5</sup>; and Phillips divides by the low-temperature limit of the Debye temperature.<sup>3</sup> None of these will bring the three curves into agreement.

The failure of the homology rule for diamond indicates that the interatomic forces must be different from those in silicon and germanium.

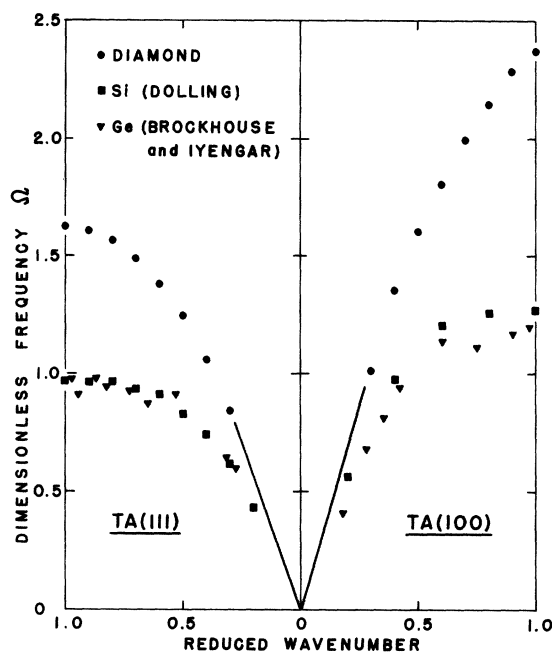


FIG. 1. Dispersion curves for diamond, silicon, and germanium in dimensionless units. If the three elements were homologous, the curves should coincide. The solid lines were calculated from the elastic constants of diamond given by McSkimin and Bond.

It may be noted that in order to explain the very flat TA dispersion curves observed in silicon and germanium, it was necessary to take account of the deformation of the electron shells of the atoms during their vibration.<sup>10</sup> This suggests that this deformation is less important in the case of diamond.

Two rather different sets of elastic constants have been reported for diamond.<sup>11,12</sup> Our data are probably not accurate enough at small values of  $Z$  (where systematic errors are likely to be largest) to decide between them. However, our present data seem to favor the results of McSkimin and Bond. We hope to be able to shed more light on this point and also to obtain some data on the higher branches of the diamond dispersion curves.

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<sup>1</sup>B. N. Brockhouse, *Inelastic Scattering of Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1961), p. 113.

<sup>2</sup>The diamond was loaned to the U. S. Atomic Energy Commission by Industrial Distributors, Ltd., a Division of DeBeers Consolidated Mines, Ltd.

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<sup>5</sup>H. Bilz, R. Geick, and K. F. Renk, *Proceedings of the International Conference on Lattice Dynamics, 1963* (to be published).  
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<sup>11</sup>S. Bhagavantam and J. Bhimasenachar, *Proc. Roy. Soc. (London)* **A187**, 381 (1946).  
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### SELF-DETECTION OF THE ac JOSEPHSON CURRENT\*

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In carrying out current-voltage ( $I$ - $V$ ) measurements on a large number of Pb-(Pb oxide)-Pb superconducting tunnel junctions<sup>1</sup> exhibiting dc Josephson currents,<sup>2-4</sup> we have observed a temperature-independent resonance-shaped peak at applied voltages  $V_0$  less than the energy gap,  $2\Delta$ . This structure is only readily apparent at low temperatures (1.2°K to 1.6°K) and when the sample is in a magnetic field large enough to

quench the dc Josephson current.<sup>5</sup> In Fig. 1 we show a typical series of  $I$ - $V$  curves taken at 1.2°K for several different values of magnetic field,<sup>6</sup> the field being oriented in the plane of the junction. For our sample geometry, such characteristic curves are generally independent of the particular orientation of the field in the junction plane. However, when the field is perpendicular to the junction, the peak completely disappears.

From a detailed analysis of  $I$ - $V$  curves such as given in Fig. 1, we have been able to obtain the following information:

(a) The voltage  $V_p$  at which the peak occurs is approximately a linear function of the applied magnetic field  $H_0$ , the ratio  $V_p/H_0$  being  $0.19 \pm 0.02$  when  $V_p$  is expressed in millivolts and  $H_0$  is expressed in gauss.

(b) The height of the peak is a strong function of magnetic field and consequently of voltage. For small voltages it varies as  $V_p^{-2}$ .

(c) The peak height for a given sample depends upon the magnitude of the zero-field dc Josephson current; the larger the Josephson current, the larger the peak. Indeed, for high-resistance samples which exhibit a dc Josephson current of only  $10^{-2}$  to  $10^{-3}$  of the maximum theoretical value, no peak is observed (at least down to 1.2°K).

The behavior described in (a) and (b) above is graphically presented in Fig. 2 where we plot, for the same sample shown in Fig. 1, the position of the peak as a function of magnetic field and the height of the peak as a function of voltage. Data are given only out to fields of order 10 gauss, corresponding to voltages of about  $6\Delta/5$ , because the peak becomes very small and badly smeared at larger fields (see Fig. 1).

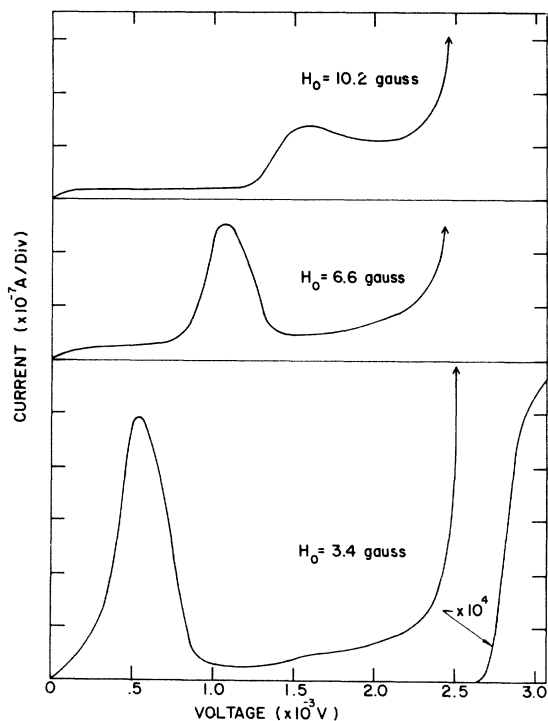


FIG. 1. Typical experimental  $I$ - $V$  curves obtained for three different values of the magnetic field.