cially at the higher scattering angles, it was difficult to place these observed "bumps" on a more definite basis.

Because of the change in the symmetry, it would also be of interest to compare the magnetic form factor of hexagonal close-packed Co with that of face-centered cubic Co. Hence we are continuing these measurements in order to refine the fcc data further and to determine the form factor of the hexagonal phase. A detailed report will be submitted upon the completion of this work.

We are indebted to Dr. T. A. Kaplan and Dr. S. J. Pickart for profitable discussions of these results. We also wish to thank Dr. H. J. Williams and Dr. R. M. Bozorth of Bell Telephone Laboratories for supplying us with the crystals.

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*This work was performed under the auspices of the U. S. Atomic Energy Commission and the National Security Agency.

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LATTICE VIBRATIONS IN SILICON AND GERMANIUM

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(Received February 13, 1959; revised manuscript received March 2, 1959)

The dispersion curves of lattice waves travelling in the [001] directions of a silicon single crystal have been measured using neutron scattering. The frequency (ν) and wave vector ($\vec{q}$) of each phonon, and the character of each branch, were determined as previously discussed.[1] The complete results for the longitudinal acoustical (LA), longitudinal optical (LO), and the degenerate transverse acoustical (TA) and transverse optical (TO) branches are shown in Fig. 1.

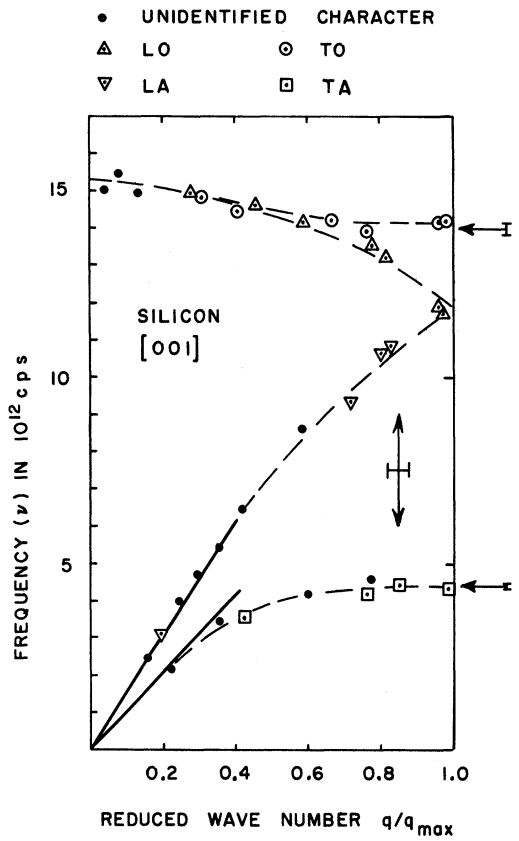

FIG. 1. Frequency vs reduced wave number for lattice vibrations propagating in the [001] direction of the reduced zone in silicon at room temperature. The heavy lines have the slope of the appropriate velocity of sound as found by ultrasonic measurements [H. J. McSkimmin, J. Appl. Phys. 24, 988 (1953)]. The vertical arrow indicates the presumed position (q_0) of the minimum of the conduction band (reference 13). The horizontal arrows indicate the frequencies of phonons at q_0 found from the indirect transition (references 10-12).

Two series of experiments were performed using a new multiple axis crystal spectrometer at the NRU reactor. In the first, neutrons of 1.900A wavelength were scattered from a single crystal of silicon at angles in the vicinity of 90° with the crystal oriented so that the wave vector of the outgoing neutrons ($\vec{k}'$) lay very nearly along the line ($\bar{2}$, $\bar{2}$, 3) to ($\bar{2}$, $\bar{2}$, 6) of the silicon reciprocal lattice.[2] The wave vectors of all phonons observed in these experiments thus lay in the [001] direction of the reduced zone. In the second series, neutrons of 1.900, 1.350, and 1.693 angstroms were used in the method of successive approximations,[1] in which phonons were measured in regions of reciprocal space in which their polarization, and optical or acoustical character, could be unambiguously determined. All measurements were made at room temperature. The points are believed to define the level of all branches to about 3%. Values of important frequencies (units 10^{12} cps) are: Raman ($q=0$) 15.3 ± 0.3, TO[001] 14.2 ± 0.3, L[001] 11.9 ± 0.5, TA[001] 4.35 ± 0.15.

The measurements are in disagreement with x-ray measurements of Learn.[3] The Raman frequency is in agreement with a prediction[4] 15.45 $\times 10^{12}$ cps based on the far infrared spectrum, and with an independent neutron measurement of 15.2×10^{12} cps by Palevsky, Hughes, Kley and Tunkelo.[5] The dispersion curves are in agreement with predictions[4,6] from specific heat, elastic constant, and far infrared measurements, that the frequencies for silicon are similar to those for germanium at similar positions in the zone but higher by a factor of about 1.75. Six figures of merit obtained from these data gave[4] an average ratio 1.745 with a standard deviation of 0.04. The average ratio of the silicon frequencies reported herein to the frequencies of germanium measured by Brockhouse and Iyengar[1] at the same positions in the reduced zone is 1.73 ± 0.04. If exact homology held between silicon and germanium, the harmonic force constants would be proportional to the inverse square of the lattice constant (a), and the frequencies therefore proportional to $(Ma^2)^{-1/2}$, where M is the mass of the atom. Thus the frequency ratio would be 1.675, slightly but significantly lower than the measured ratios. The expected frequency ratio can also be computed from the melting temperatures using the Lindemann law,[7] leading to a value of 1.96. The lack of complete homology thus seems to be connected with an abnormally low melting point in germanium (compared with silicon).

It follows from the approximate homology that in silicon, as in germanium, either long-range forces between atoms are important or the Born-Von Kármán theory is inadequate.[1,8,9].

The [001] direction was selected for initial study because the minimum of the conduction band lies in this direction, and it was hoped that the wave vector ($\vec{q}_0$) of the minimum could be located by comparison of these results with other types of measurements which are dependent on the value of $\vec{q}_0$. Measurements of the infrared absorption spectrum by Macfarlane, McLean, Quarrington, and Roberts[10] have been interpreted to give values for the frequencies at q_0 of 4.4, 14.0, 21.9, and 29.5×10^{12} cps. Measurements of the spectrum of recombination radiation by Haynes, Lax, and Flood[11] originally gave frequencies a little lower than those obtained by Macfarlane et al., but more recent measurements are in agreement.[12] The two higher frequencies of each set are clearly inconsistent with the results of Fig. 1 but could arise from double phonon processes as suggested[4] earlier. From measurements of electron nuclear double resonance, Feher[13] has found that $q_0/q_{max} = 0.85 \pm 0.03$. If this value is accepted, then the two lower frequencies of each set must be TA and probably TO. The LO and LA transitions are presumably weak and overshadowed by the very strong TO transition.

The measurements of lattice frequencies of germanium of Brockhouse and Iyengar,[1] discussed above, and those of Ghose, Palevsky, Hughes, Pelah, and Eisenhauer[14] are in disagreement for the TA and especially for the longitudinal branches near the zone boundary in the [001] direction. For L[001] Brockhouse and Iyengar quoted $(6.9 \pm 0.4) \times 10^{12}$ cps, while the corresponding frequency derived from the measurements of Ghose et al. is $(5.3 \pm 0.4) \times 10^{12}$ cps. In an effort to resolve this question we have remeasured[15] the [001] longitudinal branch in germanium using neutrons of longer wavelength (1.917A) than those used by Brockhouse and Iyengar (1.52A), and somewhat higher resolution. The two types of experiments discussed above for silicon were performed, and gave results in agreement with one another. The dispersion curve (nine points) obtained agreed with that of Brockhouse and Iyengar, the frequency of the L[001] phonon being $(7.0 \pm 0.3) \times 10^{12}$ cps.

The author wishes to thank Dr. J. R. Haynes, Dr. M. Lax, Dr. J. Burton, and Dr. M. Tanenbaum of Bell Telephone Laboratories for the loan of the crystals.

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[5] Palevsky, Hughes, Kley, and Tunkelo, this issue [Phys. Rev. Lett. 2, 258 (1959)].

[6] Similar homology arguments have been presented in the past; see references quoted in reference 4 and F. Herman (private communication).

[7] See N. F. Mott and H. Jones, Properties of Metals and Alloys (Oxford University Press, London, 1936), p. 13.

[8] F. Herman, paper given at Conference on Semiconductors (see reference 4).

[9] M. Lax, Phys. Rev. Lett. 1, 133 (1958). The arguments of Lax on silicon are still applicable even though

the actual phonon frequencies are greatly different from those he used, which were those given by the infrared workers.

[10]Macfarlane, McLean, Quarrington, and Roberts, Phys. Rev. 111, 1245 (1958); paper given at Conference on Semiconductors (see reference 4).

[11]Haynes, Lax and Flood, Bull. Amer. Phys. Soc. Ser. II, 3, 30 (1958); paper given at Conference on Semiconductors (see reference 4).

[12]J. R. Haynes (private communication).

[13]G. Feher, paper given at Conference on Semiconductors (see reference 4).

[14]Ghose, Palevsky, Hughes, Pelah, and Eisenhauer, Phys. Rev. 113, 49 (1959).

[15]These experiments were performed after the initial submittal of this Letter.

LATTICE VIBRATIONS IN SILICON BY SCATTERING OF COLD NEUTRONS*

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The use of subthermal or "cold" neutrons for investigating the lattice vibrations in solids has proved to be of increasing value in the past several years. Upon absorption of a phonon (the quantum of lattice vibration energy) while scattering, the neutron energy changes by such a large amount that the phonon energy can usually be accurately fixed. The method is particularly useful for the optical vibrations, as shown by the first measurements[1] of high-frequency modes in zirconium hydride and germanium, cases for which infrared work had given inconclusive results. In the light of similar uncertainty concerning the optical (Raman) frequency in silicon, we have determined it, as well as one acoustical branch, by use of cold-neutron scattering.

Because of the great interest in the electronic properties of silicon, a number of infrared measurements have been performed, which have been used to infer the Raman frequency, the optical vibration of zero phonon momentum. For example, Collins and Fan,[2] on the basis of infrared absorption measurements, conclude that $h\nu$ for the Raman line is 0.076 ev. Haynes, Lax, and Flood,[3] from a measurement of recombination lines from infrared excitation, obtain optical lines between 0.08 and 0.12 ev. Macfarlane, McLean, Quarrington, and Roberts,[4] on the basis of infrared absorption edge measurements, report similar values, 0.09 and 0.12 ev. Recently, Cole and Kineke[5] have reported that the unpublished x-ray results of A. Learn give a value of 0.071 ev. On the theoretical side, Hsieh,[6] using the theory of Smith,[7] has calculated the optical vibration spectrum from the measured force constants and obtained a Raman value of 0.077 ev, using only first neighbor interactions, and a value of 0.10 ev when the effects of second neighbors are included. These discordant results indicate the need for an unambiguous value of the optical line, such as that obtainable from cold-neutron scattering.

In the cold-neutron technique as developed at Brookhaven,[8] the neutron beam incident on the scatterer is produced by filtration through 8 inches of beryllium. No Bragg (elastic) scattering can occur for the low-energy (~0.005 ev) incident neutrons, and those scattered at 90° (the position of the detector) are of necessity those that have gained energy by interacting with the lattice vibrations. Since the energy of the optical vibrations of silicon is in the neighborhood of 0.05 ev, the neutrons experience a tenfold increase in energy when scattered (Fig. 1). It is this large change in energy, giving directly the phonon energy, that is a distinct advantage of the cold-neutron method.

In the present measurements, the incident neutron beam was perpendicular to the (100) plane of the silicon single crystal. The energy of neutrons scattered at 90° was determined by measuring their time of flight over a 5-meter flight path, utilizing a slow chopper. Figure 1 shows a typical set of experimental data, plotted in terms of time of flight. The sharp peak at high energy represents the absorption of one quantum of vibrational energy from an optical mode, and the one at lower energy corresponds to an acoustical mode. Data similar to Fig. 1 were obtained for a number of orientations of the crystal as it was rotated about the [100] axis. The results give energies for phonons of various momenta $\vec{q}$, with $\vec{q}$ limited to the (100) plane of reciprocal lattice space. Points for $\vec{q}$ in the [110] direction, extrapolated to $\vec{q}=0$, give the energy of the Raman line as 0.063 ± 0.003 ev (phonon frequency, ν, of 15.2×10^{12} cps); and at the edge of the Brillouin zone (maximum $\vec{q}$) in the [110] direction the transverse acoustic branch has an energy of 0.020 ± 0.002 ev ($\nu = 4.84\times10^{12}$ cps). Thus our value for the Raman line is not in agreement with the infrared measurements nor with the Hsieh calculations. It is possible, however, to reinter-