21, 1331 (1968).

[12] B. W. Faughnan and Z. J. Kiss, IEEE J. Quantum Electron. QE-5, 17 (1969).

[13] Z. J. Kiss and W. Phillips, Phys. Rev. 180, 924 (1969).

[14] L. Rimai and G. A. de Mars, Phys. Rev. 127, 702 (1962).

[15] E. S. Kirkpatrick, K. A. Müller, and R. S. Rubins, Phys. Rev. 135, A86 (1964).

[16] B. W. Faughnan (unpublished).

[17] The $SrTiO_3$:Mo crystal has some Al^{3+} added to compensate the Mo^{6+}. Otherwise, the Ti lattice will reduce, thereby producing conduction electrons. The optical absorption of the conduction electron would make the observation of the Mo^{5+} band difficult.

[18] R. S. Rubins and W. Low, in *Proceedings of the First International Conference on Paramagnetic Resonance, Jerusalem*, 1962, edited by W. Low (Academic, New York, 1963), pp. 59–67.

[19] W. Low and E. L. Offenbacher, *Solid State Physics*, Vol. 17 (Academic, New York, 1965), p. 136.

[20] K. A. Müller, W. Berlinger, and R. S. Rubins, Phys. Rev. 186, 361 (1969).

[21] See discussion of line shapes below.

[22] V. G. Bhide and H. C. Bhasin, Phys. Rev. 172, 290 (1968).

[23] B. W. Faughnan and Z. J. Kiss, Bull. Am. Phys. Soc. 12, 642 (1967).

[24] T. C. Ensign and S. E. Stokowski, Phys. Rev. B 1, 2799 (1970).

[25] K. A. Müller, Th. von Waldkirch, W. Berlinger, and B. W. Faughnan, Solid State Commun. (to be published).

[26] M. I. Cohen and R. F. Blunt, Phys. Rev. 168, 929 (1968).

[27] D. L. Dexter, *Solid State Physics*, edited by F. Seitz and D. Turnbull, Vol. 6 (Academic, New York, 1958).

[28] Manuel Cardona, Phys. Rev. 140, A651 (1965).

[29] I. Broser, K. H. Franke, and H. J. Schulz, in *Proceedings of the International Conference on Semiconductors*, edited by D. G. Thomas (Benjamin, New York, 1967), p. 81.

PHYSICAL REVIEW B VOLUME 4, NUMBER 10 15 NOVEMBER 1971

Velocities of Sound in Polycrystalline Neon[†]

R. Balzer,[*] D. S. Kupperman, and R. O. Simmons
Department of Physics and Materials Research Laboratory,
University of Illinois, Urbana, Illinois 61801

(Received 26 July 1971)

The longitudinal and transverse velocities of sound have been measured in polycrystalline specimens of neon from 18 to 24.5 K by ultrasonic pulse-echo techniques. The results are compared to values at lower temperatures obtained previously by Batchelder and co-workers and by Bezuglyi and co-workers. From these results the isothermal compressibility has been calculated. Comparisons made with various model calculations show the data to be in best agreement with the improved self-consistent model calculations of Goldman, Horton, and Klein.

Noble-gas crystals have been the subject of extensive theoretical as well as experimental investigations during the past several years. The heavier gases such as argon and krypton may be taken as almost ideal examples for studying calculations of lattice theory. A great deal of experimental data for these crystals are already available. For helium, the lattice dynamics are dominated by effects arising from large zero-point motion, affecting many of the macroscopic features of solid helium. Neon represents an intermediate case, and zero-point motion is supposed to have only an intermediate influence in the dynamical description of the solid.

For solid neon, the measured thermodynamic properties, particularly the bulk modulus, have been uncertain in the upper third of its temperature range before melting. The only elastic measurement reported above 20 K, where the most rapid temperature variation is expected to occur, is a value for the bulk modulus obtained by Batchelder and co-workers from x-ray isobaric lattice thermal expansion of crystals at pressures of 0 and 8 bar (800 kN m^{-2}), respectively.[1] At 20 K the measured relative change in lattice parameter between 8 and 0 bar was about $(4.5 \pm 0.5) \times 10^{-4}$. When one compares this to the value of the coefficient of linear thermal expansion, about 13×10^{-4} deg^{-1} near 20 K, one sees that temperature differences of order 20 mdeg could contribute the majority of the uncertainty.

Because the high-temperature region is a particularly sensitive test of lattice models, and because large experimental uncertainty exists there, further independent measurements are desirable. Polycrystalline specimens are easiest to prepare, but they exhibit considerable grain growth and suffer easy plastic deformation at high temperatures. This renders measurements somewhat variable, and the present work is no exception, but these disadvantages have been somewhat overcome here by the use of many specimens.

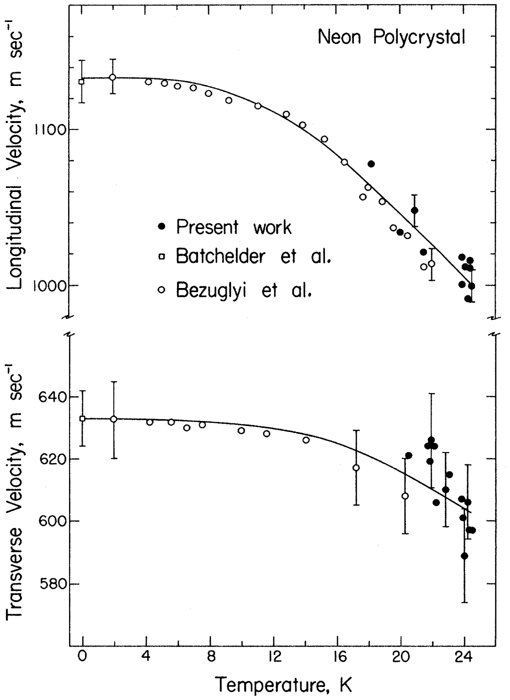

FIG. 1. Longitudinal and transverse sound velocities in polycrystalline neon with comparison to results of previous investigators. Longitudinal velocities were measured in five specimens and agree with the results of Bezuglyi *et al.* in the region of overlap. Transverse velocities were measured in eight specimens where the use of many specimens should yield an appropriate polycrystalline average.

In the present paper, measurements of the sound velocities in polycrystalline specimens of natural neon using a pulse-echo technique are described. The cryostat and method used here are described earlier for previous sound velocity measurements on solid krypton.[2] The specimens were grown from gas of 99.995% purity in a Kapton (polypyromelitimide film) tube by rapid unidirectional solidification from the melt directly on either a 10-MHz *X*-cut or *AC*-cut quartz transducer exciting longitudinal or transverse waves.

Specimens grown were examined with a 4*X* telescope and were found to be clear and dense and without any visible inhomogeneities. The average grain size was determined by thermal etching and was found to be less than 0.2 cm^2. Temperatures were measured by a calibrated germanium resistance thermometer. Individual specimen lengths, about 5 mm, were measured by a cathetometer to an estimated uncertainty of ± 0.03 mm. Transit times were measured by a calibrated oscilloscope. Up to 3 echoes were visible and the uncertainty in the transit times is estimated to be ± 0.05 μsec. Differential thermal contraction between specimen and transducer limited the range of temperature over which the velocities could be measured.

A check for possible systematic errors was carried out *in situ* on specimens for which the sound velocity is already well known: liquid neon at 26 K for the longitudinal velocity, and three specimens of polycrystalline argon in the range 78–83 K for the transverse velocity. The results were within 1% of the values presently available in the literature.[3,4]

Results for the velocity of longitudinal sound in five polycrystalline specimens of neon of somewhat different lengths and grain structures, all grown in the same specimen tube, are shown in Fig. 1. The velocities of transverse sound measured in eight polycrystalline specimens of neon, all grown in the same specimen tube and again having somewhat different lengths and grain structures, are also shown in Fig. 1. The uncertainties in these measurements are estimated to be about 1% for longitudinal waves and 2% for transverse waves.

The longitudinal and transverse velocities at 0 K were deduced by Batchelder *et al.*[1] following direct x-ray measurements of isothermal bulk modulus *B* at 4.25 K. The relation for the adiabatic bulk modulus is

$$B_s = \rho(v_l^2 - \tfrac{4}{3} v_t^2) \,, \tag{1}$$

and for the Debye temperature at 0 K

$$\Theta = \frac{h}{k}\left[\frac{4\pi M}{9N\rho}\left(\frac{1}{v_l^3} + \frac{2}{v_t^3}\right)\right]^{-1/3} \quad . \tag{2}$$

M is the gram atomic weight of natural neon ($M = 20.183$ g mole^{-1}). At 0 K one has $B_s(0) = (1.12 \pm 0.03) \times 10^9$ N m^{-2},[1] $\Theta(0) = (74.6 \pm 1.0)$ K,[5] and $\rho(0) = 1.5072$ g cm^{-3}.[1] In the temperature range between 2 and 20 K, sound velocity measurements on polycrystalline neon were

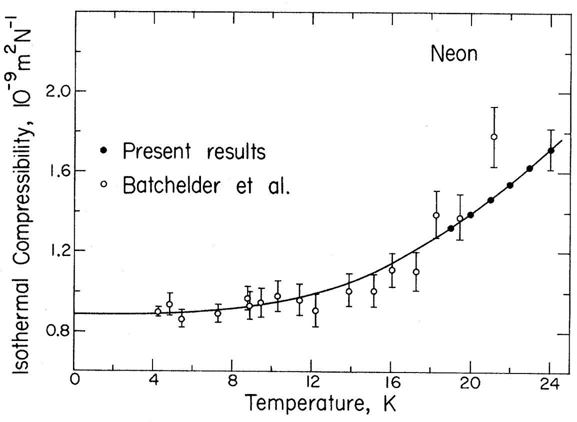

FIG. 2. Isothermal compressibility calculated from the smoothed polycrystalline velocities of sound and Eqs. (1) and (3). Comparison to the x-ray results of Batchelder *et al.* (Ref. 1), while within the limits of experimental error, shows a considerable difference in the compressibility near the triple point.

TABLE I. Smoothed values of some thermodynamic functions of polycrystalline neon versus temperature (estimated possible uncertainty in parentheses): longitudinal sound velocity v_l(± 1%), transverse sound velocity v_t(± 2%), adiabatic bulk modulus B_s(± 5%), isothermal compressibility χ_T, and x-ray density ρ.

T (K)	v_l (msec^{-1})	v_t (msec^{-1})	B_s (10^9N m^{-2})	χ_T (10^{-9}m^2 N^{-1})	ρ (g/cm^{-3})
2	1133	633	1.13	0.885	1.5073
3	1133	633	1.13	0.886	1.5072
4	1132	632	1.13	0.888	1.5072
5	1131	632	1.12	0.891	1.5070
6	1130	632	1.12	0.896	1.5068
7	1128	632	1.12	0.903	1.5063
8	1126	631	1.11	0.913	1.5056
9	1124	631	1.10	0.926	1.5045
10	1120	631	1.09	0.942	1.5032
11	1117	630	1.08	0.962	1.5015
12	1112	629	1.06	0.987	1.4994
13	1107	628	1.05	1.02	1.4968
14	1100	627	1.02	1.05	1.4939
15	1092	626	0.998	1.10	1.4905
16	1083	625	0.971	1.15	1.4867
17	1074	623	0.943	1.20	1.4825
18	1065	621	0.915	1.26	1.4779
19	1055	619	0.887	1.32	1.4728
20	1045	616	0.860	1.39	1.4672
21	1035	613	0.834	1.46	1.4610
22	1026	610	0.809	1.54	1.4543
23	1016	607	0.784	1.62	1.4470
24	1006	604	0.758	1.71	1.4394

obtained by Bezuglyi *et al.*[6] with ultrasonic waves of frequency 1 MHz (Fig. 1). The region of overlap near 20 K shows fairly good agreement with the present results. The sound velocities in solid neon are therefore given with some assurance over the entire temperature range from 0 K up to the triple point (24.55 K).

The adiabatic bulk modulus has been calculated from the smoothed polycrystalline wave velocities of Fig. 1 by means of Eq. (1). It was then converted to the isothermal compressibility from the following expression:

$$\chi_T = \frac{1}{B_s} + \frac{\beta^2 TM}{\rho C_p} , \qquad (3)$$

where values for the temperature-dependent quantities β and ρ are tabulated in Ref. 1, and C_p in Ref. 7. The temperature dependence of the isothermal compressibility obtained in this manner is shown in Fig. 2. The results of Batchelder *et al.* obtained from direct x-ray measurements are also included. The results show good agreement and are consistent within the experimental error. The values obtained by Batchelder *et al.* from isobaric thermal expansion have the highest accuracy at low temperatures, where the thermal expansion of solid neon is the smallest. Table I gives smoothed values for v_l, v_t, B_s, χ_T, and ρ from 2 K up to the triple point.

Early self-consistent harmonic calculations,[8] which exclude odd-order terms in the expansion of the crystal free energy and employ Mie-Lennard-Jones (MLJ) 12-6 potential parameters obtained from another source, do not agree well with the present experimental results. From this it appears likely that terms of the type omitted (cubic anharmonic terms) contribute significantly to zero-temperature thermodynamic and elastic properties of neon. The sound velocities derived by Gillis *et al.*[8] differ from the measured values for neon from 10 to 20% over the entire temperature range.

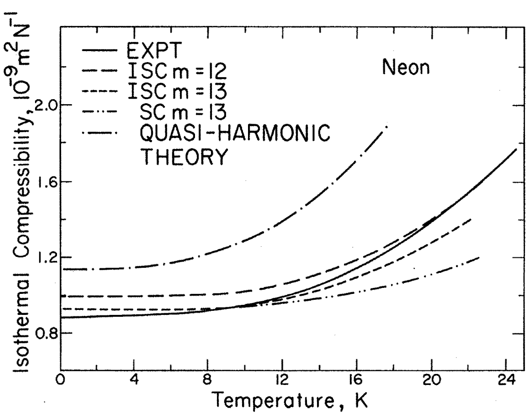

FIG. 3. Comparison of the experimental results for the isothermal compressibility with theoretical model calculations of Goldman, Horton, and Klein (Ref. 9).

The leading odd-order correction to self-consistent theory was incorporated in work by Goldman *et al.*,[9] who retained the convenient MLJ m-6 potential, but used both $m = 12$ and $m = 13$ with more appropriate potential parameters. The resulting compressibility, in a 12-6 model, is about 10% too large near 0 K but within the limits of experimental error near the triple point. For a 13-6 model, theoretical and experimental values essentially agree near 0 K but the theoretical values for compressibility fall somewhat below the limits of experimental error at temperatures close to the triple point. The comparisons are shown on Fig. 3.

In such calculations the choice of the appropriate potential is still open to some question. Present knowledge of the elastic behavior of solid neon indicates that some further work on the properties of strongly anharmonic fcc crystals is worthwhile.

[†] Work supported in part by the U. S. Atomic Energy Commission under Contract No. AT(11-1)-1198.

[*] On leave of absence from I. Phys. Inst., Techn. Hochschule Darmstadt, Germany.

[1] D. N. Batchelder, D. L. Losee, and R. O. Simmons, Phys. Rev. 162, 767 (1967).

[2] D. S. Kupperman and R. O. Simmons, J. Phys. C 4, L5 (1971).

[3] E. V. Larson, D. G. Naugle, and T. W. Adair, Phys. Letters 32A, 443 (1970).

[4] G. J. Keeler and D. N. Batchelder, J. Phys. C 3, 510 (1970).

[5] H. Fenichel and B. Serin, Phys. Rev. 142, 490 (1966).

[6] P. A. Bezuglyi, R. O. Plakhotin, and L. M. Tarasenko, Fiz. Tverd. Tela 12, 1199 (1970) [Sov. Phys. Solid State 12, 934 (1970)].

[7] W. E. Schoknecht, dissertation (University of Illinois at Urbana-Champaign, 1971) (unpublished).

[8] N. S. Gillis, N. R. Werthamer, and T. R. Koehler, Phys. Rev. 165, 951 (1968).

[9] V. V. Goldman, G. K. Horton, and M. L. Klein, J. Low Temp. Phys. 1, 391 (1969).

PHYSICAL REVIEW B VOLUME 4, NUMBER 10 15 NOVEMBER 1971

Lattice Relaxation and Small-Polaron Hopping Motion[*]

David Emin
Sandia Laboratories, Albuquerque, New Mexico 87115

(Received 22 March 1971)

The three-dimensional analog of Holstein's molecular crystal model is utilized as the basis for a study of the relaxation of the lattice after a small-polaron hop. In particular, it is shown explicitly that the time-dependent activationlike energy arising in the previously developed theory of correlated small-polaron hopping motion is directly related to the actual relaxation of the lattice from the distorted configuration it must assume to facilitate a small-polaron hop. That is, the time dependence of this "activation energy" and the concomitant relaxation of the hop-related lattice displacements are governed by a single entity denoted as the relaxation function. Furthermore it is demonstrated that this function is directly expressible in terms of the transfer of vibrational energy from the initially distorted sites to successive (initially undistorted) neighbor sites. In fact, for the most part, the relaxation of the lattice after a hop is associated with the transfer of vibrational energy to only nearest-neighbor sites, this being essentially a local phenomenon independent of the periodic nature of the lattice. Finally, although the lattice relaxation for our three-dimensional model is found to proceed much faster than in the previously developed one-dimensional model, its effect on small-polaron hopping motion may not be inconsequential. In particular, the small-polaron drift mobility is shown to be significantly affected by lattice relaxation effects when the mean time between small-polaron hops is less than or comparable to the lattice relaxation time; this time being essentially the reciprocal of the optical-phonon bandwidth parameter.

I. INTRODUCTION

The notion of small-polaron hopping motion has been advanced in order to explain the transport properties of excess carriers in a number of low-mobility materials. Specifically, in such materials the electron-lattice interaction is viewed as strong enough so as to make possible the self-trapping of a carrier. That is, the excess carrier distorts the surrounding lattice via the electron-lattice interaction thereby producing a sufficiently deep potential well so that it is bound in its own "induced trap." The unit composed of the carrier, localized with a spacial extent of the order of a lattice constant, and its induced lattice distortion is termed a small polaron.

Fundamental to our understanding of the mechanism which characterizes the hopping motion of a