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Difference between zero- and first-sound propagation in solid Kr

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We point out that the zero-sound–first-sound difference in solid rare gases is a very sensitive probe of the phonon self-energy. Recent advances in our knowledge of the interatomic potentials of these solids have enabled us to remove much of the ambiguity from this source and hence focus directly on the influence of different model self-energies. We have investigated the approximations of Koehler and Goldman, Horton, and Klein using a self-consistent harmonic basis with no explicit inclusion of hard-core effects. We find that the previously unexplained experimental result for solid Kr near melting, that C_{44} as measured by neutron scattering is 12% greater than obtained from Brillouim scattering, lies between the predictions of these two approximations.

I. INTRODUCTION

Of the many manifestations of vibrational anharmonicity in solids, one of the strangest is that the velocity of sound as measured by low-frequency-ultrasound or Brillouin-scattering experiments (so-called first sound) is different, at finite temperatures, from that measured in the high-frequency regime as measured for example in neutron-scattering experiments (zero sound). First or zero sound occurs when the frequency of the sound wave is either less than or greater than the inverse lifetime of the thermally populated phonons in the specimen being studied. This effect has been known for some time[1,2] and has been observed experimentally by Svennson and Buyers[3] in KBr, Blinick and Maris[4] in Si, and by Jackson, Landheer, and Stoicheff[5] in solid Kr. The latter authors found the value of C_{44} obtained from inelastic-neutron-scattering data[6] on solid Kr at 114 K, C_{44}^0, to be more than 12% greater than C_{44}^S obtained from their Brillouin-scattering experiments at 115.6 K. The only theoretical estimates available[7,8] suggested about a 5% effect. The purpose of this paper is to focus on approximations that go into the theoretical calculations to see if this difference is a real one, or just a manifestation of these approximations. Solid Kr is a particularly suitable solid to study because the interatomic forces between Kr atoms are now rather well understood. Bobetic and Barker[9] obtained a potential for the Kr_2 dimer which, along with the three-body Axilrod-Teller-Muto (ATM) force, gave a good over-all account of many low-temperature properties. However, the thermal expansion of the solid calculated from this model was a little too big[10,11] and moreover the dimer potential did not fit the differential-collision cross sections from crossed-molecular-beam studies.[12] This lead to refinement of the Kr_2 dimer potential and two new almost identical potentials[13,14] have appeared recently that do not suffer from the above-mentioned defects and moreover give excellent fits to gas-phase transport data.[15] The potential from Ref. 13 also gives a better fit[16] to the experimental phonon dispersion curves at 10 K of Skalyo *et al.*[17] than the earlier potential of Bobetic and Barker. It seems that the use of this potential in solid-state calculations should remove most of the ambiguity arising from uncertainty in the interatomic forces and hence enable us to concentrate on the approximations entering into the treatment of the lattice dynamics.

In Sec. II we outline the calculations we have performed and give some selected results. Anticipating our results we shall see that the effect reported by Jackson *et al.*[5] can be reconciled with uncertainties in the theoretical calculations. We end the paper with some concluding remarks about the situation in other rare-gas solids.

II. OUTLINE OF THE CALCULATION AND RESULTS

We have evaluated the elastic constants of solid Kr using the new Kr interatomic potential[13] following the calculational procedures outlined by Klein, Horton, and Goldman,[18] with a minor modification of the computer programs to incorporate the all-neighbor potential. Two approximations for the anharmonic contributions are examined due to Horner,[19] and Götze and Michel.[20] We employ a self-consistent-harmonic approximation, incorporating no explicit effects due to the hard core and treat the anharmonic terms as a perturbation. For temperatures sufficiently high enough for classical mechanics to effectively ap-

ply the exact elastic constants can be obtained by a Monte Carlo calculation.[21] A comparison of results for the elastic constants using the approximations of Horner, and Götze and Michel with the Monte Carlo calculation for the Bobetic-Barker potential[22] is given in Table I. There we see that the "exact" result lies between these two approximations, being slightly closer to the Götze-Michel approximation. The isothermal bulk modulus includes the contribution from three-body ATM forces. The elastic constants for the new potential of Ref. 13 are given in Table II for both the Horner, and Götze-Michel approximations. There are only relatively minor differences between the elastic constants of the new Barker potential[13] and the older Bobetic-Barker one.[9] The experimental Brillouin-scattering results[5] seem to lie between the admittedly rather wide bounds set by the two approximations at high temperature. At low temperature (10 K) the neutron-scattering elastic constants[17] are also in reasonable agreement with the results in Table II and this reinforces our belief in the model. The results for C_{44} are shown in Fig. 1.

We have calculated phonon energies of low wave vectors using the appropriate generalization of the approximations of Horner, and Götze and Michel to finite wave vectors.[23,24] Some results are shown in Fig. 2. Again we see that the experimental data for the branch [001]T lies between the predictions of these two approximations. The phonon energies were converted to zero-sound elastic constants and some selected values are given in Table III labeled with the superscript 0. The value of C_{44}^0 is also shown in Fig. 1.

Comparison with the adiabatic first-sound elastic constants shows that difference between zero- and first-sound elastic constant C_{44} can indeed be much larger than the previous theoretical estimates of 5%, probably lying in the (5–15)% range estimated in Table III.

Figure 1 shows that the neutron-scattering elastic constants at 10, 77, and 114 K all seem to agree closely with the Goldman-Horton-Klein (GHK) approximation for the phonon self-energy,[24] while the Brillouin-scattering result seems to lie between the approximations of Horner, and Götze and Michel.

TABLE I. Comparison of Monte Carlo (MC) calculation of the elastic constants (Ref. 22) with the results of using the phonon self-energy approximations of Horner, Ref. 19, Σ_H; and Götze and Michel, Ref. 20, Σ_{GM}. The calculations were carried out at 115 K and $a = 4.125$ Å and employ the Bobetic-Barker Kr_2 potential, Ref. 9.

	MC	Σ_H	Σ_{GM}
C_{44}	12.5± 0.3	9.78	13.47
$C_{11}-C_{12}$	8.6± 0.3	7.61	10.11
B_T+ATM	14.6± 0.5	9.1	15.2

We find evidence (see Table III) of a positive dispersion in the lowest-energy branch $[O\xi\xi]T_1$ as reported experimentally.[6,25] On the other hand, we also found it impossible to reconcile our calculations with the enormous zero-sound–first-sound difference in the bulk modulus reported by Peter *et al.*[25] for solid Kr at 77 K. Our calculated values for the transverse branches at this temperature are in good accord with their measurements. It may be that the resolution correction for the longitudinal branches employed by Peter *et al.* was somewhat larger than estimated.

III. CONCLUDING REMARKS

We have shown that the previously unexplained large differences between the zero- and first-sound elastic constants of solid Kr near its melting point can be reconciled with theoretical uncertainties in the phonon self-energy. The calculations we employ do not take explicit account of effects due to the hard core of the interatomic potential. It may well be that application of the ideas of Horner[26] along the lines of those recently attempted by Kanney and Horton[27] will help remove some of this uncertainty. A calculation using the computer-simulation molecular-dynamics method[28] may also be instructive, especially since this will

TABLE II. First-sound elastic constants in kbar using two different approximations for the phonon self energy: Σ_H (Horner, Ref. 19), Σ_{GM} (Götze-Michel, Ref. 20). The results are obtained using the Kr_2 potential from Ref. 13 and include contributions from all neighbors.

Temp. K	$(C_{11}-C_{12})$ Σ_H	$(C_{11}-C_{12})$ Σ_{GM}	C_{44} Σ_H	C_{44} Σ_{GM}	C_{11}^S Σ_{GM}	C_{11}^S Σ_{GM} +ATM	B^S+ATM Σ_{GM}
0	21.19	21.22	26.83	26.87	45.3	49.7	35.54
10	20.99	21.02	26.62	26.67	44.9	49.3	35.23
77	13.44	14.48	17.03	18.62	33.3	36.8	27.19
114	8.44	10.68	10.51	13.88	25.0	28.0	20.87
115.6	8.21	10.52	10.21	13.68	24.7	27.6	20.58

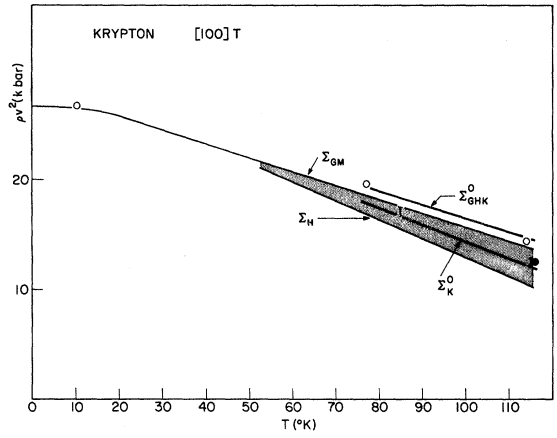

FIG. 1. Temperature dependence of C_{44} for solid Kr. The open circles are taken from experimental neutron scattering data in Refs. 6, 17, and 25. The full circle is the Brillouin-scattering result from Ref. 5. The two error bars are Monte Carlo values from Ref. 22. The solid lines labeled Σ^0_K and Σ^0_{GHK} are calculated zero-sound values for C_{44} using the approximations of Refs. 23 and 24, respectively. The curves labeled Σ_H and Σ_{GM} are the elastic constants calculated using the approximations of Refs. 19 and 20.

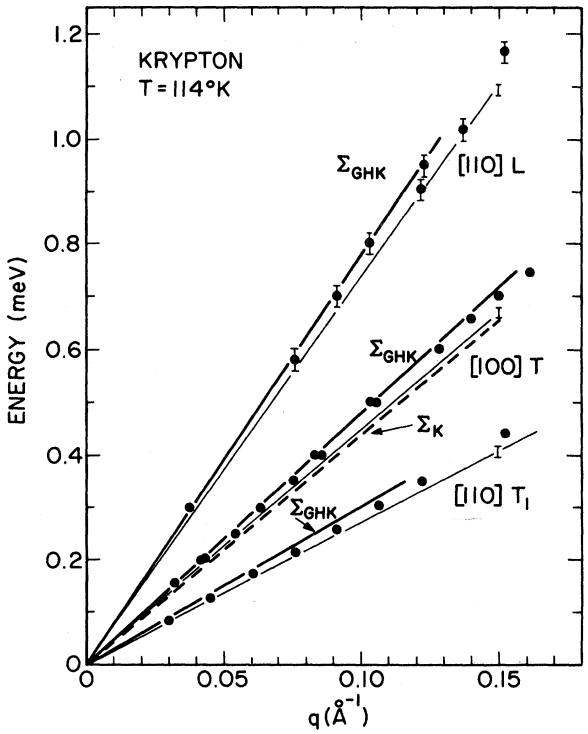

FIG. 2. Phonon dispersion curves for solid Kr at 114 K and low wave vector q. The points are the experimental neutron-scattering data from Ref. 6. The thin lines are the extrapolation of the Brillouin-scattering wave velocities from Ref. 5. The solid lines labeled Σ_{GHK} are calculated using the self-energy of Ref. 24, while the dashed line labeled Σ_K uses the approximations of Ref. 23.

give an essentially exact result for a classical solid like Kr near melting.

Similar effects to those observed in solid Kr have been reported[29-31] recently for solid Ar and Ne, while solid Xe appears to show anomalous behavior.[32-34] Further experiments on solid Xe would be particularly valuable since at present there seems to be no way to reconcile the existing data with current theoretical ideas.

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TABLE III. Comparison of C^S, adiabatic first-sound elastic constants, with C^0, the elastic constants obtained from the slopes of phonon dispersion curves using the phonon self-energy approximations Σ_{GHK} (Goldman-Horton-Klein, Ref. 24) and Σ_K (Koehler, Ref. 23). The results are obtained using the Kr_2 potential from Ref. 13 and include contributions from ten shells of neighbors.

Temp. K	ξ	Direction	Σ	C^S (kbar)	C^0 (kbar)	$\Delta = 100\frac{C^0 - C^S}{C^S}$
114	0.03	[00ξ]T	GHK	13.94	14.86	6.6
	0.10	[00ξ]T	GHK	13.94	14.65	5.1
	0.10	[00ξ]T	K	10.57	12.15	14.9
77	0.10	[00ξ]T	GHK	18.69	19.33	3.4
	0.10	[00ξ]T	K	17.10	18.03	5.4
114	0.02	[0$\xi\xi$]T_1	GHK	5.38	5.94	10.4
	0.045	[0$\xi\xi$]T_1	GHK	5.38	6.12	13.8

[1]H. J. Maris, Philos. Mag. 16, 1331 (1967).
[2]R. A. Cowley, Proc. Phys. Soc. Lond. 90, 1127 (1967).
[3]E. C. Svennson and W. J. L. Buyers, Phys. Rev. 165, 1063 (1968).
[4]J. S. Blinick and H. J. Maris, Phys. Rev. B 2, 2139 (1970).
[5]H. E. Jackson, D. Landheer, and B. P. Stoicheff, Phys. Rev. Lett. 31, 296 (1973).
[6]J. Skalyo, Jr. and Y. Endoh, Phys. Rev. B 7, 4670 (1973).
[7]V. V. Goldman, G. K. Horton, and M. L. Klein, Phys. Rev. B 4, 567 (1971).
[8]G. Niklason, Phys. Kondens. Mater. 14, 138 (1972).
[9]M. V. Bobetic and J. A. Barker, Phys. Rev. B 2, 4169 (1972).
[10]M. L. Klein, T. R. Koehler, and R. L. Gray, Phys. Rev. B 7, 1571 (1973).
[11]M. L. Klein and R. D. Murphy, Phys. Rev. B 6, 2433 (1972).
[12]M. Cavallini, M. G. Dondi, G. Scoles, and V. Valbusa, Entropie 42, 136 (1971).
[13]J. A. Barker, R. O. Watts, J. K. Lee, T. P. Schafer, and Y. T. Lee, J. Chem. Phys. 61, 3081 (1974).
[14]U. Buck, M. G. Dondi, V. Valbusa, M. L. Klein, and G. Scoles, Phys. Rev. A 8, 2409 (1973).
[15]R. Aziz, Mol. Phys. (to be published).
[16]J. A. Barker, M. V. Bobetic, and M. L. Klein (unpublished).
[17]J. Skalyo, Jr., Y. Endoh, and G. Shirane, Phys. Rev. B 9, 1797 (1974).
[18]M. L. Klein, G. K. Horton, and V. V. Goldman, Phys. Rev. B 2, 4995 (1970).
[19]H. Horner, Z. Phys. 205, 72 (1967).
[20]W. Götze and K. H. Michel, Z. Phys. 217, 170 (1968).
[21]M. L. Klein and W. G. Horner, Phys. Rev. B 4, 537 (1971).
[22]M. L. Klein and R. D. Murphy, Phys. Rev. B 6, 2433 (1972).
[23]T. R. Koehler, Phys. Rev. Lett. 22, 777 (1969).
[24]V. V. Goldman, G. K. Horton, and M. L. Klein, Phys. Rev. Lett. 24, 1424 (1970).
[25]H. Peter, J. Skalyo, Jr., H. Grimm, E. Lüscher, and P. Korpiun, J. Phys. Chem. Solids 34, 255 (1973).
[26]H. Horner, Solid State Commun. 9, 79 (1971); and Chap. 8 in *Dynamical Properties of Solids*, edited by G. K. Horton and A. A. Maradudin (North-Holland, Amsterdam, 1974).
[27]L. B. Kanney and G. K. Horton, report (unpublished).
[28]J. P. Hansen and M. L. Klein, J. Phys. Lett. 35, L-29 (1974).
[29]S. Gewurtz and B. P. Stoicheff, Phys. Rev. B 10, 3487 (1974).
[30]Y. Fujii, N. A. Lurie, R. Pynn, and G. Shirane, Phys. Rev. B 10, 3647 (1974).
[31]R. A. McLaren, H. Kiefte, D. Landheer, and B. P. Stoicheff, Phys. Rev. B 11, 1705 (1975).
[32]Y. Endoh, G. Shirane, and J. Skalyo, Jr., Phys. Rev. B 11, 1681 (1975).
[33]N. S. Gornall and B. P. Stoicheff, Phys. Rev. B 4, 4518 (1971).
[34]N. A. Lurie, G. Shirane, and J. Skalyo, Jr., Phys. Rev. B 9, 2661 (1974).