Two-phonon scattering in solid neon

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The dynamic form factor $S(Q, \omega)$ appearing in the cross section for inelastic neutron scattering from phonons in solid neon at 5 K, is calculated using the self-consistent phonon theory. Comparison of the computed $S(Q, \omega)$ with the scattering intensities observed by Skalyo et al. shows that the unusually shaped groups they found for high-frequency phonons in neon is due to an unusually large and frequency-dependent two-phonon scattering component.

I. INTRODUCTION

Some time ago, Skalyo et al.¹ made an interesting and detailed study of the dynamics of solid neon by means of inelastic neutron scattering. In the course of the study, unusual and unexpected shapes of the dynamic form factor, $S(Q, \omega)$, were observed when scattering from high-energy longitudinal phonons having wave vector $\vec{q}$ near the zone boundary was attempted. Usually, the part of $S(Q, \omega)$ which corresponds to scattering from a single phonon can be clearly identified as a sharp Lorentzian-shaped peak superimposed on a uniform background due to scattering from multiples of phonons. However, for high-frequency longitudinal phonons having wave vectors along the [100] direction in neon, Skalyo et al. found the one-phonon peak had a large shoulder on the high-frequency side as shown in Fig. 1. Explanations of this shape have been proposed in terms of scattering from two phonons,¹ unusual intrinsic one-phonon line shapes, and in terms of interference between one-phonon and multiphonon scattering.² The purpose of this work is to present a calculation of $S(Q, \omega)$ which reproduces these line shapes accurately and thereby to identify the origin of the shoulder. Eckert et al.³ have recently experienced a similar difficulty in identifying $S(Q, \omega)$ for high-frequency phonons in fcc ⁴He at low temperature.

In Sec. II we outline how $S(Q, \omega)$ was calculated here, present the results for the phonon groups in Sec. III, and discuss them in Sec. IV.

II. DYNAMIC FORM FACTOR

$S(Q, \omega)$ for solids is usually expanded in a power series of $(\langle \vec{Q} \cdot \vec{u} \rangle)^p$, where $\vec{u}$ is the vibrational displacement of an atom from its lattice point and $\vec{Q}$ is the wave vector transferred in the scattering process. This expansion expresses $S(Q, \omega)$ as a sum,⁴⁵

$$S(Q, \omega) = S_1(Q, \omega) + S_{12}(Q, \omega) + S_{2s}(Q, \omega) + \cdots$$

$$= S_{1}(Q, \omega) + S_{2}(Q, \omega) + \cdots$$

corresponding to scattering from single phonons ($S_1$), from pairs of phonons ($S_{12}$), and so on. In this expansion we have also included the interference term ($S_{12}$) between the one- and two-phonon scattering contributions. The observed $S(Q, \omega)$ may be used to study the single-phonon properties of a crystal provided $S_1(Q, \omega)$ can be clearly identified in the total sum of all scattering processes. This is usually the case since $S_1(Q, \omega)$ has a strong resonance behavior peaking when $\omega \approx \omega(q, \lambda)$, where $\omega(q, \lambda)$ is the frequency of the single phonon having reduced wave vector $\vec{q} = \vec{Q} - \vec{T}$ and branch $\lambda$ involved in the scattering.

FIG. 1. Scattering intensity from [100] $L$ phonons with incident neutron energy $E_p = 35$ meV observed by Skalyo et al. The instrument resolution was $\approx 1.7$ meV for $\xi \approx 0.7$. The upper curves are fitted to two Gaussians where the integrated intensity of the left-most peak has been fixed.

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The two-phonon part of $S_2(Q, \omega)$, for phonons having infinite lifetime, takes the form

$$S_2(Q, \omega) = \frac{1}{N} \sum_{i=1}^{N} f_1^2(Q \cdot \epsilon_i) f_2^2(Q \cdot \epsilon_i) \times J(1, 2, \omega) \Delta(Q - (\epsilon_i + \epsilon_j)),$$

where $J(1, 2, \omega)$ is the two-phonon response function

$$J(1, 2, \omega) = (n_1 + n_2 + 1) \pi \delta(\omega - (\omega_1 + \omega_2))$$

$$- \delta(\omega + \omega_1 + \omega_2)$$

$$+ (n_2 - n_1) \pi \delta(\omega + (\omega_2 + \omega_1))$$

$$- \delta(\omega - \omega_2 + \omega_1)).$$

Here $\omega_1$, $\epsilon_i$, and $q_i$ are the frequency, polarization vector, and wave vector of one of the phonons involved in the scattering, respectively, $n_i = \kappa(\omega_i)$ is the Bose function, $d(q)$ is the Debye Waller factor and $f_1^2 = h/2 M \omega_1$, where $M$ is the atomic mass. Since $J(1, 2, \omega)$ includes a sum over all phonons in the crystal, $S_2(Q, \omega)$ is usually a flat uniform function of $\omega$. In this case $S_2(Q, \omega)$ will be a peaked function superimposed on a flat background due to scattering from two, three, and higher multiples of phonons. The properties of the interference term, $S^{II}(Q, \omega)$, have been discussed extensively elsewhere.4,7,8

To calculate $S(Q, \omega)$ for solid neon at $5$ K and lattice constant $a_0 = 4.454$ Å we have used the self-consistent phonon theory9 including cubic anharmonic corrections.10 Here the cubic anharmonic term leads to phonon damping in $S_1(Q, \omega)$. The coupling between one and two phonons through the cubic term also makes interference possible between the one- and two-phonon parts of $S(Q, \omega)$. The self-consistent harmonic frequencies without damping were used to calculate $S_2(Q, \omega)$. The Ne-Ne interaction was represented by the Goldman-Klein11 Morse–Van der Waals spline function.

### III. PHONON GROUPS

The calculated $S(Q, \omega)$ shown in Fig. 2 reproduces the observed scattering intensity of Fig. 1 well, including particularly the shoulder on the high-frequency side of the one-phonon peak.18 This shoulder is the two-phonon scattering contribution $S_2(Q, \omega)$ which has a large frequency dependence. $S_2(Q, \omega)$ is small at small $\omega$, rises rapidly in the frequency range $h \omega \sim 6-8$ meV and at higher $h \omega$ dominates $S_1(Q, \omega)$ as can be seen from Fig. 3 where $S_1$ and $S_2$ are shown separately. The difference between $S_2$ and $S_2$ also shown in Fig. 3 shows that the interference terms are not important in determining the overall shape of $S(Q, \omega)$.
The rapid frequency dependence of $S_{\nu}(\vec{Q},\omega)$ arises since the neon crystal is cold, $T \ll \theta_D \approx 75\,\text{K}$. In this case all $\kappa(\omega) = 0$ and only the first term in $J(1,2,\omega)$ contributes for $\hbar \omega > 0$. This first term will be small until $\omega$ reaches the sum $\omega_1 + \omega_2$, for two phonons both of which come from toward the end of a dispersion curve where the density of phonon states is high. The lowest transverse branch in neon has an energy $\lesssim 3.1\,\text{meV}$ so we expect $J(1,2,\omega)$ to be very small for $\hbar \omega \lesssim 6\,\text{meV}$. Thereafter $J(1,2,\omega)$ will rise rapidly as the $\delta$-function condition becomes satisfied for a large number of phonons. To test this argument, we calculated $S_{\nu}(\vec{Q},\omega)$ for an artificial neon crystal at 75 K. At this temperature $S_{\nu}(\vec{Q},\omega)$ is uniform over all $\omega$ having a slight peak at low $\hbar \omega$ in agreement with the group shapes found, for example, by Hansen and Klein in a molecular-dynamics study of a high-temperature Lennard-Jones solid.

IV. DISCUSSION

We find that at low temperature $S_{\nu}(\vec{Q},\omega)$ has approximately the same general frequency dependence as shown in Fig. 3 for all values of $\vec{Q}$. Thus, the same type of two-phonon background will accompany all one-phonon components $S_{\nu}(\vec{Q},\omega)$ independent of the reduced wave vector $\vec{q} = \vec{Q} - \vec{F}$ of the one phonon selected. Hence, for those selected phonons of low frequency, where the one-phonon function peaks below 6 meV in neon, the $S_{\nu}(\vec{Q},\omega)$ will be small and the one-phonon part readily identifiable. However, for all phonons of energy $\gtrsim 6\,\text{meV}$ the one-phonon group will lie just in the rapidly frequency-dependent region of $S_{\nu}(\vec{Q},\omega)$ and will be difficult or impossible to separate from the two-phonon scattering. In particular the peak of $S_{\nu}(\vec{Q},\omega) + S_{\nu}(\vec{Q},\omega)$ in Fig. 3 lies at higher energy and is broader than that of $S_{\nu}(\vec{Q},\omega)$ alone which could lead to error in identifying one-phonon frequencies and lifetimes.

This character of $S_{\nu}(\vec{Q},\omega)$ is common to all fcc crystals of atoms having a large zero-point vibrational amplitude which interact via a pairwise potential having a step repulsive core. Eckert et al.\textsuperscript{3} for example, have recently experienced a similar difficulty in measuring the frequencies and lifetimes of all high-energy phonons in fcc $^4\text{He}$ at low temperature. The high-energy $\{100\}$ phonons in Ar observed by Egger et al.\textsuperscript{14} and Batchelder et al.\textsuperscript{15} appear to show an artificial shift to high energy, but not the more recent and accurate data of Fuji et al.\textsuperscript{16} nor that of Skalyo et al.\textsuperscript{17} in Kr. The smaller zero-point vibration presumably reduces $S_{\nu}(\vec{Q},\omega)$ in these heavier-rare-gas crystals.

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\begin{thebibliography}{99}
\bibitem{glyde1974} P. F. Choquard, \textit{The Anharmonic Crystal} (Benjamin, New York, 1965).
\bibitem{eckert1972} Strictly, in the constant-incoming-neutron-energy method used by Skalyo et al., the scattering intensity is not quite proportional to $S(\vec{Q},\omega)$. Rather the observed intensity is $\sim 25\%$ lower on the high-energy-transfer ($\hbar \omega$) side relative to $S(\vec{Q},\omega)$ in Fig. 1 so that the height of the shoulder would be somewhat increased if a correction were made.
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