[9]As will be described elsewhere, antiperiodic conditions are particularly useful for calculating the "helicity modulus" or, for a Bose fluid, the superfluid density $\rho_s(T)$.

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[15]We assume a zero ordering field ζ. Nonzero values can be included by allowing X^T to depend also on the standard thermodynamic scaling variable $y=\zeta/t^{\Delta}$, where $\Delta=\beta+\gamma=\beta\delta$.

[16]The proposal (18), but without the condition $\lambda>1$, was first advanced by P. G. Watson [J. Phys. C: Proc. Phys. Soc., London 1, 268 (1968)], on less general grounds. Watson (private communication) has since withdrawn his claim that it is correct for a spherical model.

X-Ray Brillouin Scattering

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An investigation of x-ray scattering, when the Bragg condition is nearly satisfied, reveals the possibility of studying phonon phenomena at momenta as small as 10^4 cm^{-1}. A closer study of elastic Bragg scattering in that region provides an explanation for the previously observed "coherent crystal radiation."

In the past, x-ray and neutron investigations[1-3] of phonon phenomena have only been extended to momentum transfers as small as 10^6 to 10^7 cm^{-1}, which is about 100 times larger than that measured by laser techniques. In the x-ray case, it was expected that a large background produced by elastic Bragg scattering, which was known to be experimentally indistinguishable on the basis of energy considerations from the inelastic thermal diffuse scattering (TDS), would obscure the signal. In the neutron case, the finite energy and angular resolution typically employed in neutron scattering together with the large specimens required also restricted the range of investigation to relatively large values.

In this work, we show how a closer study of elastic and TDS components when the Bragg condition is nearly satisfied (i.e., when the momentum transfer is almost equal to a reciprocal-lattice vector $\vec{G}$) shows that the two components have different phase-matching conditions. Experimentally, when we investigated phenomena at $\vec{G}\pm\vec{q}$, where $\vec{q}$ was on the order of 10^4 cm^{-1}, the two components were easily separated by using a triple-crystal spectrometer. The region of q space that can be investigated depends primarily upon the perfection of the crystals, the degree of input collimation, and not on the intrinsic width of the Bragg peak. In addition, these studies provide an explanation for the previously reported "coherent crystal radiation"[4] and have ramifications for the general field of x-ray diffraction.

We consider the situation where a well-defined (infinitely collimated) beam $\vec{K}_{in}$ is incident on a perfect crystal. The crystal has its surface cut perpendicular to a reciprocal-lattice vector $\vec{G}$. Neglecting the small phonon energy and refraction effects, the basic equations governing both Bragg scattering and TDS are

$$\vec{K}_{out}-\vec{K}_{in}=\vec{G}+\vec{q}\,,\qquad |\vec{K}_{in}|=|\vec{K}_{out}|=K\,. \tag{1}$$

Here $\vec{K}_{out}$ is the scattered-photon's wave vector and $\vec{q}$ is either the spread in G associated with the interaction effects described by dynamical[5,6] theory, or a phonon wave vector. We are interested in the region where the angular deviation of the input δ_{in} and the output δ_{out} beams from the Bragg angle θ_B are small. The angles are defined by the following relations[7]:

$$\begin{aligned} &|\vec{G}|=2|K|\sin\theta_B\,,\\ &\vec{K}_{in}\cdot\vec{G}=-|K||\vec{G}|\sin(\theta_B+\delta_{in})\,,\\ &\vec{K}_{out}\cdot\vec{G}=|K||\vec{G}|\sin(\theta_B+\delta_{out})\,,\\ &\vec{G}\cdot\vec{q}=|\vec{G}||\vec{q}|\cos\theta_q\,. \end{aligned} \tag{2}$$

All the vectors are in the same plane (Fig. 1).

As a result of the strong interaction of the x

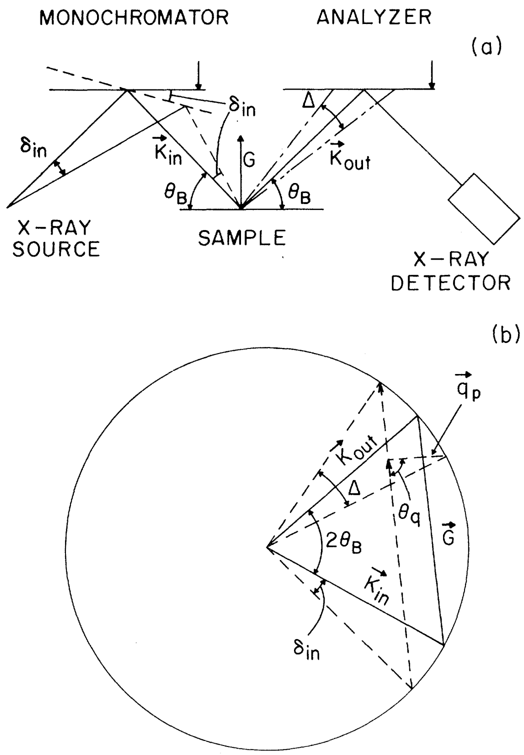

FIG. 1. (a) Schematic of the x-ray triple-crystal spectrometer in the parallel mode [1, −1, 1]. (b) Phase matching (Ewald) circle for the sample.

rays with the crystalline media,[5,6] the Bragg scattering process for a perfect crystal exists over a relatively large range of angles. The basic shape of the reflection curve is a region of perfect reflectivity centered on the Bragg angle with slowly decaying tails on either side. The angular width of the high-reflectivity region, rocking-curve width, and the extent of the tails depend in detail upon the strength of the Bragg process compared with other competing processes such as photoelectric absorption.

What has not been fully appreciated until now is that the requirement of the equality of the tangential components of the x-ray electric field at the crystal surface together with the energy-conservation condition ($|K_{in}| = |K_{out}|$) require that, for Bragg scattering, $\vec{q}$ must be perpendicular to the surface. If $\vec{G}$ is also perpendicular to the surface, then $\cos\theta_q$ equals ± 1, and Eq. (1) specifies that the reflection is specular (i.e., $\delta_{in} = \delta_{out}$). Thus, a well-defined input beam incident on a perfect crystal will undergo Bragg scattering into an equally well-collimated output beam. The width of the Bragg-scattering curve simply means that the crystal acts as a partially reflecting mirror over a range of angles. In practice, the angular width of a Bragg-scattered beam from a crystal, oriented so that it is in the reflecting region, only depends upon the collimation of the input beam and the perfection of the crystal (the spread in $\vec{G}$).

For TDS, the situation is quite different. There is no limitation on the direction or magnitude of q because there are phonons in all directions and all wavelengths. In spite of this, the TDS for a highly collimated input beam peaks at a specific output direction. The important factors for this study are that in the temperature region $KT > \hbar\omega$, the occupation of a given mode varies like $1/\omega$, and the amplitude of a given mode with unit occupation varies like $1/\sqrt{\omega}$. Therefore, TDS, which is proportional to the square of the displacement [occupation × (amplitude)^2], varies like $1/\omega^2$. For small q ($\omega = V_s q$, where V_s is the velocity of sound), TDS will vary as $1/q^2$ and thus peak where q is a minimum. By solving Eqs. (1) and (2) for the magnitude of q in terms of θ_B, θ_q, and δ_{in}, one finds that the condition for the minimum q is $\theta_q = \pi/2 - \theta_B$. This requirement and Eqs. (1) and (2) lead to

$$\delta_{out} = -\delta_{in}\cos 2\theta_B\,. \qquad (3)$$

Note that for $\theta_B < 45°$, the TDS peaks at an angle δ_{out} which is *opposite* in direction from where Bragg scattering peaks for a given δ_{in}. The width of TDS is quite large and depends in detail upon the phonon density of states.

For a given δ_{in}, the difference in output directions should be given by

$$\Delta = \delta_{out}^{Bragg} - \delta_{out}^{TDS} = 2\delta_{in}\cos^2\theta_B\,. \qquad (4)$$

If one represents the effective angular width of the Bragg reflection due to input collimation and crystal perfection by Δ_c, then the two peaks (Bragg and TDS) should be clearly separable when $\Delta \geq 2\Delta_c$. Using Eqs. (1), (2), and (4), one finds that for TDS the minimum q one can study is given by

$$|\vec{q}|_{min} = 2K\Delta_c \tan\theta_B\,, \qquad (5)$$

so that for $\Delta_c = 10$ sec, $\delta_{in} = 14$ sec, $\theta_B = 20°$, and $\lambda = 1.54$ Å, one should be able to see phonons with q's of 10^4 cm^{-1}.

To experimentally investigate these ideas, we have used a triple-crystal spectrometer.[8] The spectrometer was operated primarily in the parallel [1, −1, 1] configuration illustrated in Fig. 1(a). By also using the dispersive [1, −1, 1] configuration, we were able to verify that no energy

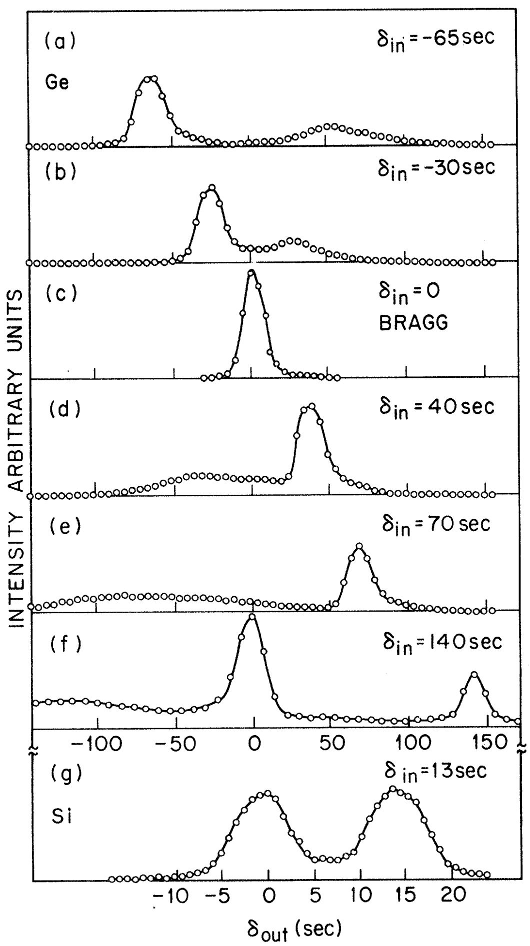

FIG. 2. Spectra obtained by sweeping the analyzer crystal on either side of the aligned Bragg position. (a)-(e) Spectra for different values of δ_{in} when the sample and analyzer were flat [220] germanium crystals and the monochromator was a grooved [220] germanium crystal. The sharp lines are due to Bragg scattering and the broad bands to TDS. (f) Spectrum with the grooved [220] germanium monochromator replaced by a flat [220] germanium crystal. Note the presence of the extra component. (g) Spectrum when all three samples were flat [220] silicon crystals. Note the change in the scale for δ_{out} and the absence of TDS because of its relative weakness.

loss larger than 1 eV occurred in any of the observed scattering phenomena. A standard point-focus copper x-ray tube was used. Germanium, silicon, and diamond crystals were studied. For the former two, we examined both [220] and [111] reflections with several surface treatments. The basic conclusions of this investigation were unaffected by the surface condition.

To achieve a highly collimated beam incident upon the second crystal (sample), the usual flat first crystal (monochromator) was replaced by a grooved germanium [220] crystal.[9] In Figs. 2(a)-2(e) are shown the scattered spectra observed by sweeping the third crystal (analyzer) for various values of δ_{in}. δ_{in} was obtained by setting the first crystal off the Bragg condition ($\delta_{in}=0$). Figure 2(c) corresponds to the Bragg setting, and one clearly sees that for small values of δ_{in} two peaks appear and move away from the old Bragg direction ($\delta_{out}=0$) as δ_{in} increases. The sharp peak which has essentially the width of the input collimation is the Bragg peak, while the broad flat peak is TDS. Our data for the [220] and [111] reflections for germanium and silicon show that the dispersion relationships $\delta_{in}=\delta_{out}$ for Bragg scattering and Eq. (4) for TDS are obeyed to within 5% for values of δ_{in} ranging from 10 to 1000 sec. The intensity of both the Bragg and TDS components decrease roughly like δ_{in}^{-2} as expected. For small δ_{in} (40 sec), we observed at the TDS peak 200 counts/sec with the x-ray tube set at 30 kV and 10 mA. In Fig. 2(b), the TDS peak was caused by phonons with wave vectors of 5×10^4 cm^{-1}. The asymmetry in the shape of the TDS peaks depending on the sign of δ_{in} is not completely understood, but the sharper one corresponds to phonons coming from the interior of the crystal to the surface while the broader one corresponds to phonons coming from the surface into the sample.

When one substitutes the grooved monochromator with a flat Ge [220] crystal, an additional peak is observed which has the same position and shape as the aligned Bragg peak and whose position and shape are independent of δ_{in}. This is shown in Fig. 2(f). Because the input to the monochromator is not collimated, its Bragg-scattered beam has a long tail. Thus, when one sets the monochromator δ_{in} from the Bragg direction, a portion of the tail is still incident upon the sample in the Bragg direction and is consequently reflected by the sample at the Bragg angle[8] ($\delta_{out}=0$). The use of a grooved crystal, which drastically reduces the tails, eliminates this component.

Studies of Ge [111], Si [110], and Si [111] yielded results identical to those illustrated for Ge [220], except for the case of Si where the TDS component is much weaker, as expected. To remove the possiblity that the observed peaks were instrumental in origin, we replaced the third crystal and detector by film and observed the three components.

As pointed out previously, the boundary conditions on the tangential component of the x-ray electric field result in $\delta_{in} = \delta_{out}$ for the Bragg component when the surface normal is parallel to G. However, a close inspection of Eqs. (1) and (2) shows that if the surface normal is not parallel to G, the $\delta_{in} = \delta_{out}$ relationship is broken quite drastically. For example, if G is 3° off from the surface normal in the plane of scattering for Si [110] or Ge [110], then $\delta_{out} = 0.7\delta_{in}$. We have observed this effect experimentally.

For the case of three flat crystals, as one reduces δ_{in} to the order of the rocking-curve width, the two Bragg components dominate, and an asymmetric profile centered near the Bragg position is observed [Fig. 2(f)]. In the dispersive [1,1,1] configuration we repeated Das Gupta and Welch's observations.[4] It seems clear from that work and the current investigation that "coherent crystal radiation" in Das Gupta and Welch's work was the "Bragg" scattering of the peak portion of the $K\alpha_1$ spectrum from the second crystal characterized by the requirement $\delta_{in} = \delta_{out}$.

Therefore this work supports the conclusion of Das Gupta and Welch that previous measurements of x-ray–emission linewidths are questionable. This skepticism should be extended to other phenomena studied by x-ray double-crystal spectrometers. We also note that the ability to measure Bragg scattering in regions in which one is not limited to a reciprocal-lattice vector and in which one does not have to worry about extinction effects raises some interesting possibilities for obtaining more accurate charge intensities by x-ray diffraction.

In this work, we have shown that investigating phonons with q's near the center of the zone is feasible. The main limitation in the technique discussed here is the perfection of the crystal. By using a microfocus x-ray system, one can work with crystals as small as 10^{-5} cm^3 and so the requirement for perfection is not so severe as it is in neutron scattering where typically 1-cm^3 crystals are required. However, x-ray scattering suffers from the defect that the energy of the phonons causing the scattering cannot be measured directly. Indirect schemes similar to those already used to determine phonon frequencies from x-ray measurements could, in principle, be applied to the case of small-q phonons. In addition to these schemes, current advances in phonon physics[10] offer the possibility of generating phonons of a known frequency into a crystal. Those schemes together with the detection technique described here offer the exciting possibility of being able to do phonon studies in many systems in a region not accessible previously. We are currently performing such measurements.

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