Observation of Surface Phonons in Inelastic Scattering of He Atoms from LiF(001) Crystal Surfaces

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Preliminary results are reported on the inelastic scattering of low-energy (20 meV), monoenergetic ($\Delta v/v = 0.8\%$ full width at half maximum) He nozzle beams from a LiF(001) surface along the ⟨100⟩ direction. The time-of-flight spectra reveal both broad and extremely sharp maxima. The broad maxima are attributed to a distribution of phonons of bulklike character with small momenta, while the sharp maxima indicate excitation of single discrete surface modes.

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Recent efforts to explore surface phonons have used the scattering of light neutral atoms and molecules.[1,2] Because of their strong interaction with the surface atoms and the fact that their kinetic energy is comparable to that of the phonons, molecular beams interact most strongly with phonons at the surface, providing a uniquely sensitive probe for investigating surface modes.

Initial attempts to detect phonon effects concentrated on the study of secondary maxima in the vicinity of diffraction peaks observed in scattered angular distributions, with no attempt to analyze the scattered-beam velocities.[3,4] Unfortunately, it does not appear possible to find a unique explanation of these secondary maxima.[5] Hence energy analysis of the scattered particles appears to be a prerequisite for unambiguous interpretation of scattering results.

In the first attempt of this sort, Bledsoe and Fisher,[6] scattering a He nozzle beam ($\Delta v/v \cong 6\%$) from LiF(001), observed but were unable to resolve "multimode" time-of-flight (TOF) peaks in the scattered beam. More recently Horne and Miller,[7] also using a He nozzle beam ($\Delta v/v = 15\%$), were able to observe a distinct elastic TOF peak, a factor of 10^2 to 10^3 lower in intensity than the specular scattering. Horne and Miller deduced the presence of a single Rayleigh surface mode with sound speed of around 4000 m/sec. Very recently we have been informed of similar experiments by Feuerbacher, Adriaens, and Thuis.[8]

In the present work the most significant difference from previous experiments is in the use of very high-speed-ratio supersonic He nozzle beams. These beams, recently developed[9] and theoretically studied[10] in this laboratory, provide a velocity resolution ($\Delta v/v < 1\%$) sufficient to resolve the predicted structure in the theoretically calculated density of states in alkali halide surfaces.[11,12]

The apparatus consits of a beam source, a target chamber, and a differentially pumped mass spectrometer detector at a fixed angle of 90° to the incident beam. The beam is chopped prior to impingement on the surface. Chopper-target and target-detector distances are 0.746 and 0.849 m, respectively. Typical measuring times are one to two hours per TOF spectra. Standard single-slot chopper TOF techniques are employed.

The present spectra were taken with the beam source cooled by liquid nitrogen ($E_{\rm He} \cong 20$ meV) and the target at room temperature. The incident and elastically scattered beams were narrowly distributed both in velocity ($\Delta v/v = 0.8\%$) and angle ($\Delta\theta_i = 0.35°$). The LiF crystal[13] was typically prepared by heating to a temperature of more than 800 °K for at least 12 h.[14] Presence of sharp diffraction peaks was taken as evidence of surface cleanliness.[1-3]

A TOF spectrum is shown in Fig. 1. Two distinct inelastic peaks are seen as well as a third which, althrough nearly buried in noise at this incident angle, dominates the spectrum near $\theta_i = 45°$.

The kinematics of single-phonon inelastic scattering are described by the equations of conservation of energy and momentum:

$$k_f^2 = k_i^2 + 2m\omega/\hbar, \qquad (1)$$

$$\Delta\vec{K} = \vec{K}_f - \vec{K}_i = \vec{G}_{m,n} + \vec{Q}. \qquad (2)$$

Here, in standard notation, $\vec{k}_i$ ($\vec{k}_f$) is the wave vector of incident (scattered) particles and $\vec{G}_{m,n}$ is the reciprocal-lattice vector, Capital letters denote components in the plane of the surface. The surface phonon has wave vector $\vec{Q}$ and frequency ω. We have thus taken $\omega < 0$ ($\omega > 0$) to correspond to creation (annihilation) of a phonon. Likewise $\vec{Q} < 0$ ($\vec{Q} > 0$) denotes a loss (gain) of transverse momentum of the incident beam particle. Only $\Delta\vec{K}$ need lie in the plane of scattering;

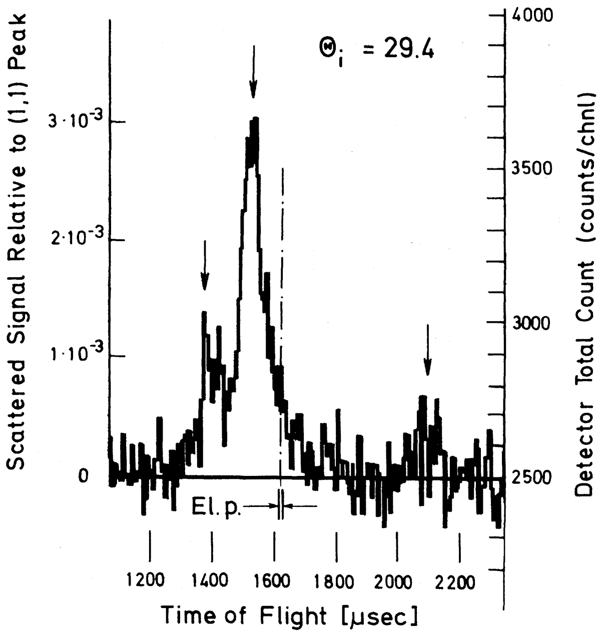

FIG. 1. TOF spectrum for the inelastic scattering of He from the LiF surface (6.9 μsec channel width, 152 min counting time). The flight time and FWHM of the elastically scattered peak (not present at this incident angle) are indicated.

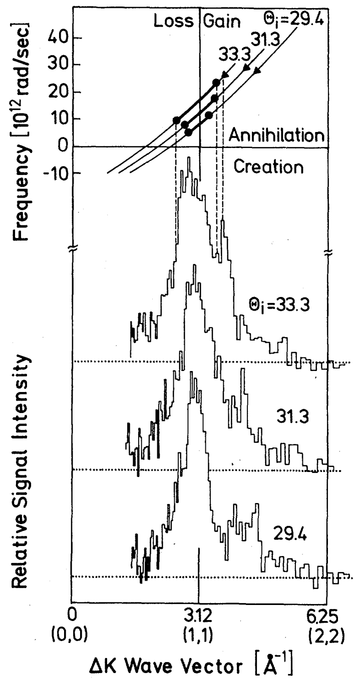

FIG. 2. Bottom: Three typical transformed TOF spectra showing broad maxima near the center of the second ($\Delta K = 3.12$ Å^{-1}) unit cell and sharp maxima to the right of the main peak. A second broad maximum near $\Delta K = 0$ has not been depicted here. Top: Section of $\omega, \Delta K$ space with common abscissa to the lower spectra. The scan curves from energy and momentum conservation are drawn and projections thereon of the features of the lower spectra are indicated (see text).

$\vec{Q}$ and $\vec{G}_{m,n}$ might have out-of-plane orientations. Combining Eqs. (1) and (2) and taking into account the fixed 90° angle then yields an equation giving the accessible values of ω and ΔK at any particular incident angle and incident beam energy:

$$\omega = \frac{\hbar^2 k_i^2}{2m}\left[\left(\frac{\Delta K}{k_i \cos\theta_i} + \tan\theta_i\right)^2 - 1\right]. \qquad (3)$$

Each TOF spectrum thus provides information on phonon density along a specific parabolic scan line through $\omega, \Delta K$ space defined by Eq. (3). To display this we have chosen to project the TOF spectra onto the ΔK axis.

Three transformed spectra are shown in the lower section of Fig. 2. The bottom of these ($\theta_i = 29.4°$) is that from the TOF trace of Fig. 1. These three spectra are representative of more than thirty spectra taken at incident angles ranging from 24° [the location of the (1,1) peak] to 45° (specular peak) on two different crystals. The dominant features are seen to be a broad maximum in the vicinity of the (1,1) reciprocal-lattice rod ($\Delta K = 3.12$ Å^{-1}) and a sharp secondary maximum at somewhat larger values of ΔK.

At the top of Fig. 2 is plotted a section of $\omega, \Delta K$ space along an abscissa common to the lower three spectra. The line curves are the parabolas from Eq. (3) for each of the three transformed spectra. Projection of each spectra upwards onto its parabola thus provides information on phonon density along the parabola. This is illustrated for the spectrum taken at 33.3°. The projection of the half width of the broad main peak is shown as a darkened curve segment and that of the sharp neighboring peak has been marked with a solid triangle. Figure 3 summarizes all the data.

Basically three factors determine the amplitude distribution of the transformed TOF spectra: (1) the density of surface modes at each point in $\omega, \Delta K$ space along the scan curve, (2) the temperature-dependent population of these modes, and (3) their cross section for interaction with the beam particles. The dominant feature of the present spectra is seen to be a broad maximum with a width which increases with increasing ω. Theoretical calculations by Benedek[11] and by Chen, de Wette, and Alldredge[12] indicate that surface-phonon dispersion relations include broad "bulk-

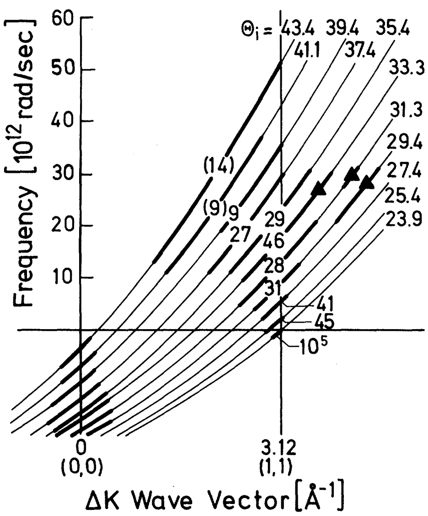

FIG. 3. Summary of results, indicating regions of dispersion space contributing to inelastic events. The FWHM's of the observed peaks are given as darkened curve segments accompanied by a number showing the peak location and its intensity relative to the (1, 1) diffraction peak. The peak locations along the upper two scan curves (parentheses) could not be definitely fixed due to decreasing signal intensity at these large ω. Triangles indicate the locations of sharp neighboring peaks as discussed in text.

like" bands. Such bands may well give rise to these maxima. The shift in location of the maxima towards momentum loss processes with increasing ω may reflect the greater thermal population of phonons with momentum loss compared to momentum gain along a specific scan curve.[8] This Bose-Einstein distribution is further responsible for the gradual decrease in peak height with increasing incident angle. Counter to this trend is the small maximum seen at $\theta_i = 33.3°$. This enhancement in inelastic scattering amplitude was also seen in the angular distribution and may be due to selective adsorption via the (1,1) reciprocal-lattice vector,[15] "kinematical focusing" of Rayleigh phonons,[16] or critical points in phonon density,[17] all of which are possible in this region.

The most striking aspect of the spectra and that indicative of the actual resolution of the apparatus is the present of sharp peaks at $\Delta K > 3.12$ $Å^{-1}$. Such features strongly suggest interactions with single, discrete surface modes even though the criterion developed by Weare[17] would indicate that the present experiments lie only on the borderline of single-phonon dominance ($\beta = 0.002$ to 0.004). That some degree of multiphonon scatter- is present would help explain the overly broad flanks of the $\Delta K = 3.12$ maxima. Such spreading may also rise from the inaccurate aximuthal alignment of the incident beam, a matter of concern with the present target manipulator, which does not allow adjustment of the azimuthal angle. The sharp peaks lie in the region of the incident angles where the above mentioned selective adsorption, kinematical focusing, or phonon critical points may enter. The preliminary nature of these data, however, does not allow us to judge the relative importance of these effects.

In summary, the present results provide convincing evidence of the potential for measuring surface modes through high-resolution inelastic beam scattering. In contrast to previous studies, several distinct peaks are seen per TOF spectrum, in accordance with the several possibilities of phonon annihilation or creation and the loss or gain in incident particle momentum. The location of the peaks shows that it is possible to observe phonons with wave vectors out to the Brillouin-zone boundary and involving nonzero reciprocal-lattice vectors. And the fact that sharp peaks are seen implies the observation of single, discrete surface modes. Further measurements are in preparation with an improved target manipulator allowing azimuthal rotation and cooling of the LiF crystal.

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Bose Condensation of Idealized Spin-Polarized Atomic Hydrogen in Equilibrium

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A model of spin-polarized hydrogen (H↑) is treated in which interactions between atoms are neglected while the single-atom Zeeman and hyperfine interactions are treated exactly. These magnetic terms in the Hamiltonian are found to affect substantially the Bose-Einstein condensation and the various thermodynamic variables. Computations are discussed of the condensation temperature, condensate density, and specific heat in order to indicate how changes in magnetic field strength might be expected to affect future measurements on this quantum system.

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Spin-polarized hydrogen (H↑) was recently stabilized in very dilute amounts[1]; if it can be produced in bulk amounts, it will be an extremely interesting quantum system. Hydrogen atoms, kept from recombining into the molecular state by a strong external magnetic field, interact via the very weak ${}^3\Sigma_u{}^+$ pair potential. The parameter $\eta=\hbar^2/m\sigma^2\epsilon$, which measures the "quantumness" of a Lennard-Jones system (with σ and ϵ the potential parameters), is 0.55 for H↑ whereas for ^{3}He it is 0.24. The high interest in this substance has resulted in numerous theoretical treatments[2-10] and some preliminary experimental studies.[11] The recent report[1] that small amounts of H↑ have been maintained without decay for long periods of time gives one strong reason to believe that bulk amounts will soon be available. H↑ is expected[4,7] to remain a Bose gas all the way to absolute zero, undergo a Bose-Einstein condensation (BEC), and presumably exhibit superfluid properties.

A BEC might be expected to manifest itself in a specific-heat peak, however, that in itself might not be enough to identify it unambiguously. Walraven and Silvera[10] have suggested that, in the nonuniform magnetic field configuration used by their group, a BEC will result in a characteristic density profile. The purpose of this paper is to identify other signatures of the BEC, which are dependent on the presence of the external field.

The present approach to the study of the H↑ BEC involves several simplifications. First, since an anlaysis involving the fully interacting Bose system is difficult, a more preliminary approach which neglects interatomic interactions is used here. However, the spin states of a single hydrogen atom in an external field are treated exactly. That is, the Zeeman and hyperfine interactions are included. Second, the external field used in the Silvera-Walraven experiment on H↑ was nonuniform so that down electron-spin states would be drawn in but up spins would be repelled. The field used in the present study is uniform but a portion of its effect is simulated in most of our calculations by simply eliminating the "wrong" electron-spin states from the energy spectrum used. Differences, of course, remain from the real situation. The condensation in a nonuniform field is a *spatial* BEC[10,12] while the one considered here is in momentum space. Thus it is expected that the present analysis will yield only a qualitative view of real H↑ which, however, may be useful in guiding experimenters in their search for effects characteristic of the BEC.

The computations reported below give results for the critical temperature T_c, the condensate fraction n_0/N, and the heat capacity at constant volume. Perhaps the most interesting results are those concerning T_c and n_0/N as a function of external field B. T_c is found to increase with in-

