

tion of the expected differences between zero- and first-sound elastic constants (and velocities) and indicate that the frequency regions of the experiments 25 to 250 GHz and 1.6 to 4.4 GHz are in or near the zero- and first-sound regimes, respectively. The large magnitude of the zero- and first-sound differences in solid krypton near the triple point, however, remains to be explained.

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Reflection of ^4He Atomic Beams from Clean LiF Surfaces*

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We have studied reflection of a ^4He nozzle beam from a LiF crystal surface cleaved *in situ* at 10^{-9} Torr. As compared with previous results on water-covered surfaces, the selective adsorption intensity minima are pronouncedly sharpened and deepened and there is considerable fine structure. The main minima can be associated with three bound states and two positive-energy resonances. On aging the surface in the vacuum, the fine structure fades and the bound-state energy levels shift slightly.

Atomic-beam scattering from the $[100]$ cleavage plane of LiF has been extensively studied in the past.¹⁻⁶ These studies were made at pressures of 10^{-8} Torr or higher on surfaces that had been prepared in various ways such as cleaving in air followed by heating in the vacuum,⁴⁻⁶ cleaving in a stream of nitrogen,³ and cleaving in the vacuum.² As O'Keefe *et al.*³ and others have pointed out, there is strong evidence⁷ that all these surfaces were covered by several monolayers of water.

We have studied the scattering of ^4He beams from LiF cleaved *in situ* at about 10^{-9} Torr, the background gases being mainly H_2 and CO . A schematic diagram of the apparatus is shown in Fig. 1. The beam has a velocity spread of about 10% and an angular spread of about 0.1 deg, with a diameter of $\frac{3}{4}$ mm at the sample. The crystal, a rod 5 mm \times 5 mm \times 50 mm, can be advanced along its length and cleaved repeatedly without breaking the vacuum. The crystal was initially γ

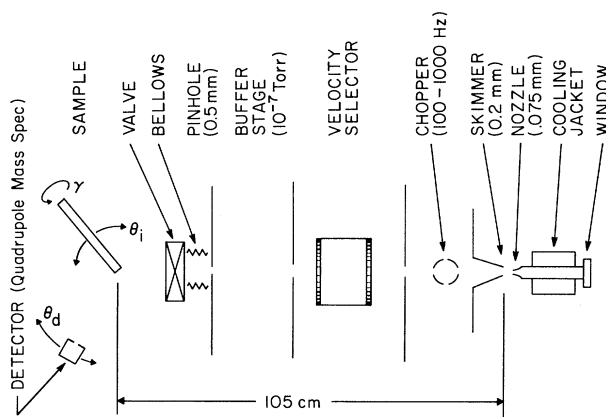


FIG. 1. Schematic diagram of apparatus. The nozzle exhaust chamber is pumped by a 600-liter/sec diffusion pump, the three subsequent chambers by 50-liter/sec diffusion pumps, and the scattering chamber by a 260-liter/sec turbomolecular pump backed by a 50-liter/sec diffusion pump.

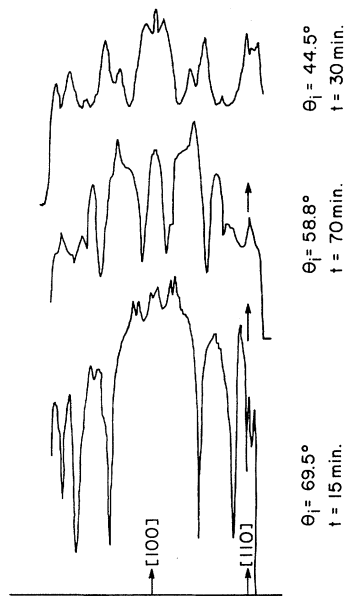


FIG. 2. Recorder tracings of specular beam intensity versus azimuthal angle γ at fixed angles of incidence. The age of the sample after cleaving is indicated on each. Maximum intensity is of the order of 30% of the incident beam.

irradiated to enhance cleavability. After baking the system at 250°C it was observed that the color centers had been bleached out, but the crystal still cleaved readily, though by no means perfectly.

The most striking difference from previous results occurs for the "selective adsorption" minima in the specularly reflected beam intensity as function of the azimuthal angle γ . Some scans made within two hours after cleaving are illustrated in Fig. 2. It is seen that the principal minima are pronouncedly deeper and sharper than was previously observed, and that there is considerable "fine structure" in the patterns. The curves shown are tracings of strip-chart recordings of the detector output made as the azimuthal angle was continuously driven (about 8 deg/min) by a clock motor. The lack of perfect symmetry in the patterns is due to imperfections in the drive mechanism and to the texture of the cleaved surface. These surfaces are decidedly rough on the optical microscope scale. Nevertheless, strong specular reflection of the helium beam (about 20 to 30% of the incident beam intensity) occurred, and similar rotation patterns were found after each of several cleavings. These data were obtained with the source chamber cooled with liquid nitrogen. A few preliminary runs with the source

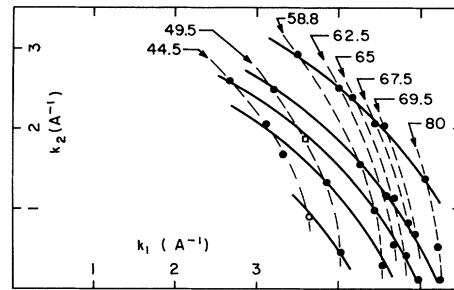


FIG. 3. Fit of main intensity minima by Eq. (1). The broken lines connect points at the same angle of incidence, indicated by the label (degrees from the normal).

at room temperature showed generally less structure.

The wavelength of the cooled beam, measured with the velocity selector (Fig. 1) and checked by the $(\bar{1}, \bar{1})$ diffraction beam, was $1.18 \pm 0.02 \text{ \AA}$, which corresponds to full expansion from a source at a temperature of about 90 K. Using this wavelength, the principal minima have been indexed according to the theory of Lennard-Jones and Devonshire^{8,9} and others.¹⁰⁻¹² This consists in fitting the angular positions of a given minimum by an equation of the form

$$\left(k_1 + \frac{2\pi}{a}n_1\right)^2 + \left(k_2 + \frac{2\pi}{a}n_2\right)^2 = k^2 - \frac{2mE}{\hbar^2}, \quad (1)$$

where \vec{k} is the incident wave vector, a is the length (2.84 Å) of the base vectors of the simple square surface mesh oriented along the $\{110\}$ directions, and E is the energy associated with motion perpendicular to the surface. The curve fitting is shown in Fig. 3. Here all the main minima have been plotted as solid dots. One group of six points and one group of seven each correspond to a minimum that can be followed over a considerable range of angles of incidence. Each of these groups falls quite unambiguously on a circle of the form of Eq. (1) with center at $n_1 = 0$, $n_2 = 1$ (the uppermost two solid curves in Fig. 3). The fitting of the remaining ten solid points is somewhat more conjectural. Various sets of circles can be drawn but none can plausibly account for all the points. The set shown in Fig. 3 has the following virtues: (a) A minimum number of points (two) are left unaccounted for; (b) the same values of n_1 and n_2 occur, whereas all other sets involve appreciably higher values; (c) no point falls on more than one circle; and (d) on reexamination of the recorder traces, distinct features can be found at the "missing" points, indicated by open symbols in Fig. 3.

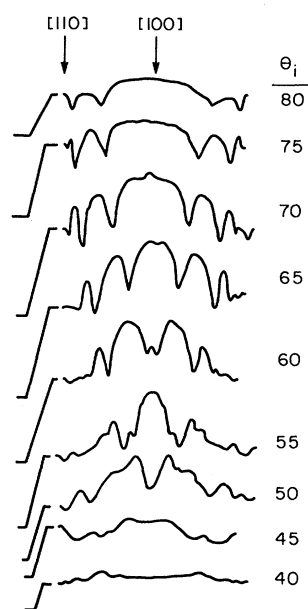


FIG. 4. Recorder tracings of specular beam intensity versus azimuthal angle γ at fixed angles of incidence. The surface had aged for three days at about 10^{-9} Torr. Maximum intensity is of the order of 5% of the incident beam.

Specifically the circle corresponds to a slight dip and the square to the middle of the "split" minimum seen in Fig. 2. The energies corresponding to the circles shown are listed in Table I, and compared with previous data. It is seen that the two deepest levels are in good agreement with previous results. In addition, a third bound state and two resonances in the unbound continuum appear. The third level is in fair agreement with calculations^{10, 13} of its expected position.

On aging the surfaces in the vacuum there was a gradual "fading" of the structure, discernible within a few hours. A set of traces made after three days is shown in Fig. 4. All the fine structure has gone, but the minima corresponding to the bound states remain clearly evident. The energies appear to have shifted somewhat deeper, to -5.9 , -3.2 , and -1.4 meV, respectively. The energies are uncertain to several tenths of an meV, because of inaccuracies in both the wavelength and the curve fitting.

Some of the fine-structure features on the fresh surfaces appear to be connected with the emer-

TABLE I. Energies of ^4He on LiF (in meV).

| Present work | Previous experiment | | | Calculation | |
|--------------|---------------------|--------|--------|-------------|---------|
| | Ref. 9 | Ref. 2 | Ref. 3 | Ref. 13 | Ref. 10 |
| -5.6 | -5.6 | -5.8 | -5.1 | | |
| -2.25 | -2.5 | | | | |
| -0.84 | | | | -0.61 | -1.1 |
| | | | | | -0.57 |
| | | | | | -0.13 |
| +0.5 | | | | | |
| +2.6 | | | | | |

gence and disappearance of diffraction beams. However, no detailed analysis has yet been attempted.

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