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Selective-Adsorption-Induced Intensity Maxima in ^{4}He/LiF Scattering

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Selective-adsorption transitions have in the past been identified with minima in the intensity of specular reflection of atomic beams. New observations show that maxima can also occur, in excellent agreement with the recent theory of Weare and co-workers.

The term "selective adsorption"[1] denotes the process in which a free particle makes a transition into one of the Bloch states of the two-dimensionally periodic potential at a crystal surface. The particle (usually an atom or molecule) is then in a bound state with respect to its motion perpendicular to the surface. Selective adsorption, as observed in molecular-beam scattering, thus offers a unique opportunity to measure the energies of such states, and thereby to gain information about the interaction potential. For this reason, it is important to have an unambiguous identification of the signature of this process as manifested in the scattering pattern.

Early theories of elastic beam-surface scattering predicted maxima in the specular [i.e., (0,0) beam] intensity. On the other hand, experiment always showed minima, and the latter were at first attributed to the supposed dominance of inelastic processes. However, Chow and Thompson[2] showed that minima could be predicted purely elastically, provided that the correct scattering channels were included in the calculation. At the same time they predicted two other effects, namely (1) maxima in certain diffracted beam intensities and (2) splitting of the specular minima due to the two-dimensional band structure. The experimental observation of these effects[3-6] lent strong support to their theory.

However, Wolfe and Weare[7] found, via a perturbation-theoretic calculation, that only the strongly coupled transitions (i.e., those induced by large Fourier components of the periodic potential) produced minima. The weakly coupled ones, they predicted, would give maxima. Indications of these effects were also seen in some of the earlier work.[2] A recent detailed calculation by Harvie and Weare[8] was compared with some experimental data obtained in this laboratory several years ago,[9] which did show indications of maxima at about the right positions.

In view of the importance of the effect, it was decided to repeat the experiment, making use of a number of technical improvements that had been incorporated in the interim. These include (1) better mechanisms for setting and reading the crystal angles; (2) cooling of the crystal to about 125 K; and (3) narrower velocity distribution (about 2% full width at half-maximum) and better stability of the ^{4}He beam. The latter is now characterized by a wave vector of magnitude $k = 5.76$ Å^{-1} and is very highly collimated to a full width of about 5×10^{-4} radian in the plane of incidence.

The sample was a bar about 6 mm×6 mm×50 mm (initial length) of "x-ray grade" LiF obtained from the Harshaw Chemical Co. This grade of material is deliberately left with a few hundred parts per million of OH^- impurity to promote hardness and cleavability. The earlier work had been done on a purer grade of material, which was hardened by γ irradiation. In all runs the crystal was cleaved while cold (125 K) and, of course, kept cold throughout the measurements.

Figure 1 is an azimuthal scan of the specular intensity, taken point by point at $\frac{1}{3}$-degree intervals at a nominal polar angle of 70°. Because of facetting of the cleavage surface, the determination of the exact polar angle involves a lengthy procedure[10] which was not carried out in the present case. Instead, the various prominent features were tentatively indexed on the basis of the nominal θ value and of the energy levels most recently determined.[10] The results are indicated in the figure by vertical lines giving the calculated unperturbed positions for the designated transitions.

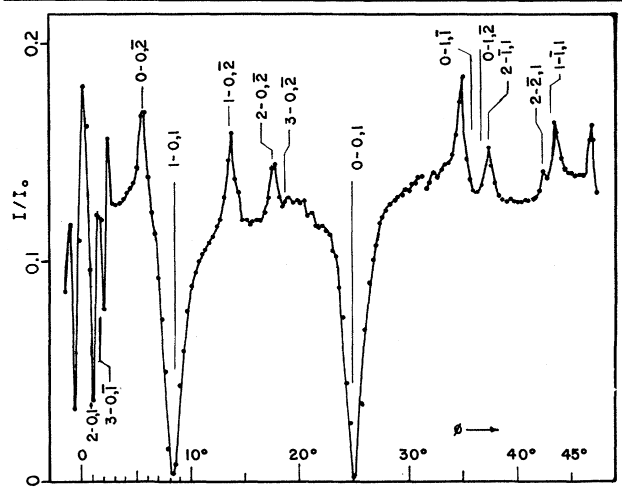

FIG. 1. Specular intensity (normalized to incident intensity) for ^{4}He ($\lambda = 1.09$ Å) on [100] cleavage face of LiF at 125 K. Polar angle 70° (nominal value). (The break at $\varphi = 30°$ is due to readjustment of the detector position, which was done every 5° to compensate for possible facetting.) The vertical marks are calculated positions of selective-adsorption transitions without correction for band-structure splittings.

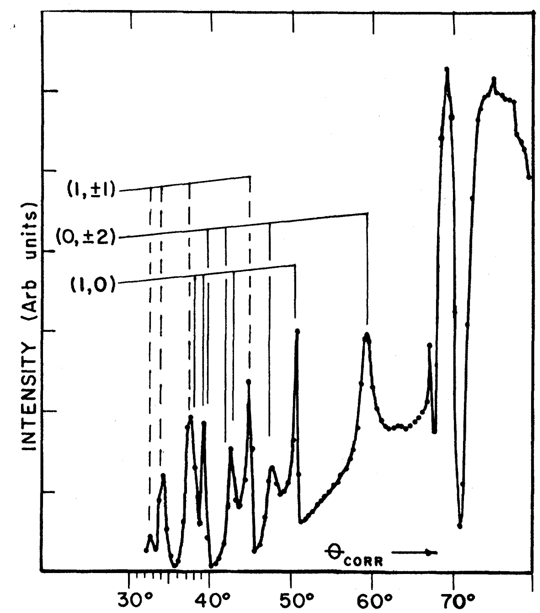

FIG. 3. Polar scan of specular intensity at $\varphi = 0$. The nominal θ values have been corrected by $-0.8°$. Vertical marks are calculated positions for transitions noted, with energy-level quantum numbers 0, 1, 2, 3 increasing from right to left.

The thermal contraction of the lattice (about 1%) was taken into account, making $g = 2.23$ Å^{-1}. It is seen that all the features fit very well except the two maxima at $\varphi = 35°$ and 37.3°. These, however, are close to the crossing of 0-(1, $\bar{1}$) and 0-(1, $\bar{2}$), which would be strongly split by the V_{01} Fourier component of the potential.

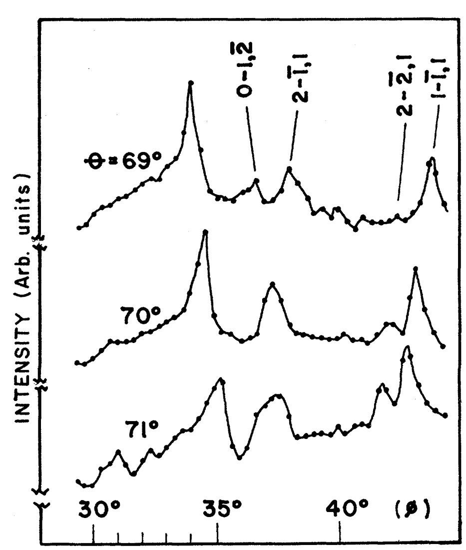

FIG. 2. Variation of specular intensity scans with polar angle. The index lines indicate expected directions of change of azimuth for the transitions noted.

The experimental results are in striking agreement with the Harvie-Weare[8] calculation. In comparing, it must be born in mind that the calculation is for a slightly shorter wavelength (1.03 rather than 1.09 Å). The main point of difference is the extent of the splitting just discussed; the calculation gave about 1.2° whereas the experiment gave about twice as much. The observed intensity is lower than that calculated, by about half, but this is, of course, expected in view of the inevitable surface imperfection.

In order to be certain of the indexing of the peaks for $\varphi > 30°$ this section of the scan was repeated for polar angles of (nomially) 69°, 70°, and 71°. These results are shown in Fig. 2. It is clear that the movements are in the expected directions. In fact for the levels not perturbed by band splitting the maxima fall very closely on the calculated circles. Thus it is confirmed that the weaker transitions do give specular maxima.

Another type of maximum is seen in Fig. 3, which is a polar scan at $\varphi = 0°$ (the [1, 1, 0] crystallographic direction) taken on a separate cleave. The two sharp minima near glancing incidence are again (0, 1) transitions. As θ is decreased, they are followed by three interleaved sets of maxima. After correcting the nominal θ values for a shift of $-0.8°$, these index quite accurately as the $(0,2)$ and $(0,\bar{2})$, $(1,1)$ and $(1,\bar{1})$, and $(1,0)$ transitions. To verify these assignments, the section of the curve around $\theta = 50°$ was repeated for $\varphi = 3°$ and $6°$. The K-plane plot of the results is shown in Fig. 4. The motions and splittings are just as expected.

Now, in these features the $(0,\pm 2)$ and $(1,\pm 1)$ transitions are weak, so that maxima are expected according to the preceding discussion. But the $(1,0)$ transitions are strong, so the maxima seem anomalous. However, Professor Weare has informed us[11] that such maxima are also predicted in cases where no open channel other than $(0,0)$ is strongly coupled to the resonant state. (A channel $\vec{G}$ is said to be "open" if $|\vec{K}+\vec{G}| < k$, where $\vec{K}$ is the surface-parallel part of the incident wave vector.) In the present case, the $(1,\bar{1})$ channel, which would be coupled to the $(1,0)$ state by V_{10}, is closed at $\varphi = 0$ for all the energy levels. As φ approaches 45°, this channel is open at all the $(1,0)$ resonances with higher energy levels, and nearly open at the (0-1,0) resonance. This probably explains why the $(1,0)$ transitions give minima near $\varphi = 45°$. The $(0,1)$ resonances, on the other hand, find the $(\bar{1},1)$ channel open (or nearly open) at all azimuths up to 45°, thus giving rise to the familiar strong minima.[9,10]

In summary, the new experimental results are in excellent agreement with and lend very strong support to Weare's theory. The new results provide yet another means of assessing the strength of the Fourier components of the potential, in addition to the band structure and multiple-scattering effects already known.

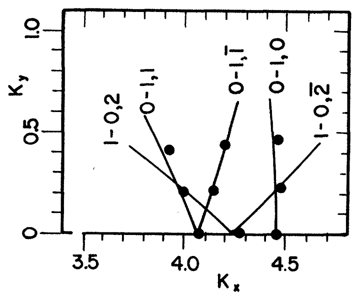

FIG. 4. K-plane plot of positions of maxima. Lines are calculated circles for transitions noted. θ values again corrected by $-0.8°$.

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