has not yet been experimentally confirmed, our study gives important experimental support to the existence of hexatic ordering. Furthermore, there exists the possibility that films of 65OBC will exhibit a 2D hexatic phase when studied in the thin film limit (2 molecular layers). These experiments are currently in progress. We note that the crossover from 3D to 2D crystalline order has already been observed in thin films of 4O.8 in its crystalline *B* phase.[1] Finally, we have also used the free-standing film technique to study smectic phases in which the molecules are tilted with respect to the layer planes. Our results[15] are in agreement with previous studies[16] which indicated a new phase without long-range positional order at temperatures below the smectic-*C* phase (tilted analogue of the smectic-*A* phase). In this case of tilted molecules, the coupling of the tilt direction to sixfold bond-orientational order adds a new and potentially interesting aspect to the physics.

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Inelastic Atom Scattering from a Cu(001) Surface and an Ordered Adsorbed Layer of Xe Atoms at 16°K

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Energy-transfer events have been observed between a He atom and a Cu(001) surface at 16°K. Only phonon creation events were observed which were confined to the subspecular region. Parallel momentum was conserved and bulk and surface modes detected. For an ordered Xe layer, both phonon creation and annihilation events were detected and parallel momentum was not conserved.

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Neutral-atom (e.g., He and Ne) scattering has recently[1,2] been demonstrated as a useful tool in detecting surface modes of vibration by measuring energy changes with use of time-of-flight techniques. Here is described a similar study with use of low-energy (22.6 meV) He atoms scattered from a Cu(001) surface at low temperature (16°K) as well as from an ordered adsorbed layer of Xe atoms deposited under various conditions. This is a desirable region for study but in the

case of metal surfaces poses experimental difficulties. The method used to observe energy changes employed a LiF analyzer crystal, the diffraction pattern from which was monitored on rotating the crystal for a fixed detector location.

The experimental arrangement was similar to that used previously[3,4] and which failed to detect inelastic events although an elastic component was observed removed from the (00) peak. Some improvements were introduced to enhance detection: (1) A rectangular beam shape (divergence of 1°) was used to increase beam intensity while maintaining similar resolution in the scattering plane; (2) the geometry was changed to place the analyzer at 25° from the Cu(001) surface when the (00) peak enters the analyzer (moved from near-normal position); (3) the detector sensitivity was increased; and (4) pumping speed in the nozzle-skimmer region has been increased. On cooling the nozzle with liquid N_2, a resolution of 1 meV was attained [full width at half maximum (FWHM) of the (11) LiF diffraction peak] at an incident energy of 22.6 meV. The resolution was not as good as that attained by Toennies and coworkers[1] and the difference was attributed to the slender skimmer design used in that case leading to a narrower incident-energy spread.

The copper crystal was prepared by a chemical cloth polishing technique (cupric chloride in hydrochloric acid) and annealed in hydrogen gas at ~1170 °K. In the beam apparatus, the crystal (aligned with ⟨100⟩ in scattering plane) was cleaned by argon-ion bombardment (600 V, 0.3 μA) and annealed at temperatures up to 600 °K for 40 min. The deposition of Xe atoms was accomplished by leaking gas via the ion beam path, thereby forming a crude atomic beam. Lower inert gases than Xe did not stick under these conditions even with the Cu crystal at the lowest temperatures. The condition of the Cu crystal was monitored by scattering a He beam into a separate detector.

Figure 1 shows the results for measured energy changes with use of the geometry shown in the inset. The analyzer direction was fixed at 130° with respect to the incident beam, i.e., $\varphi_0 + \varphi_s = 50°$, where φ_0 and φ_s are the glancing angles of incidence and scattering, respectively. If the Cu crystal was tilted so that $\varphi_0 < 25°$, the supraspecular region was accessible for energy analysis and if tilted at angles $\varphi_0 > 25°$, the subspecular region could be analyzed. The latter situation is depicted by the reciprocal lattice shown in Fig. 1. It was expected that the energy gain and loss events

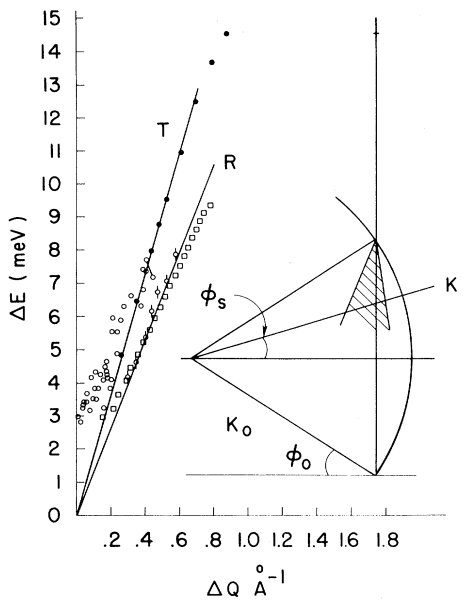

FIG. 1. The measured energy-loss changes as a function of parallel-momentum change for He scattering from Cu(001). *Inset:* The reciprocal lattice representing the geometrical arrangement of incident beam, sample crystal, and analyzer. k_0 and k are the incident- and scattered-wave vectors, respectively. φ_0 and φ_s are the glancing angles of incidence and scattering.

would be separated into the supraspecular and subspecular regions, respectively. The incident energy was always fixed (k_0 fixed), but the magnitude of the scattering vector k could be varied (or preselected) by altering the analyzer tilt angle. The target analyzer detector angle was fixed at 79.5° and 18° was the tilt angle between the (00) peak and the (11) diffraction peak which is to be located at the detector. By either varying or fixing the parameters φ_0, φ_s, and k, several types of scans in reciprocal space could be performed as will be shown below. The hatched region shown is bounded by the condition of momentum gain and loss of Rayleigh surface waves and estimated from bulk elastic constants of Cu. All other one-phonon events (resulting from bulk modes) should be detected as the locus of the scattering vector moves inside the bounded region. Any elastic events would be detected when k terminates at the sphere of reflection. A similar region occurs for the supraspecular condition should energy-gain events be detected.

Figure 2 shows scans made by fixing the Cu crystal at various values of φ_0 ($\varphi_0 > 25°$) and scanning the analyzer crystal (k varied for fixed φ_0). For each scan, the peaks on the right-hand side

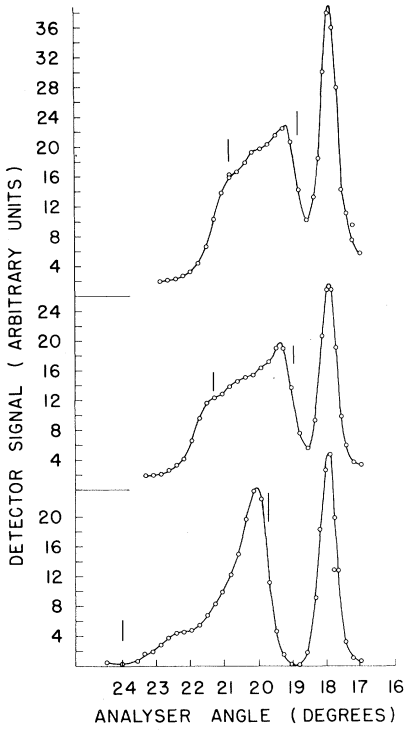

FIG. 2. Scans made by tilting the analyzer crystal, i.e., changing the tilt for fixed incidence angles from top to bottom of 29.3°, 30.1°, and 33.3°. The peaks on the RHS are the elastic component. The "band" on the LHS shows the inelastic phonon-creation component.

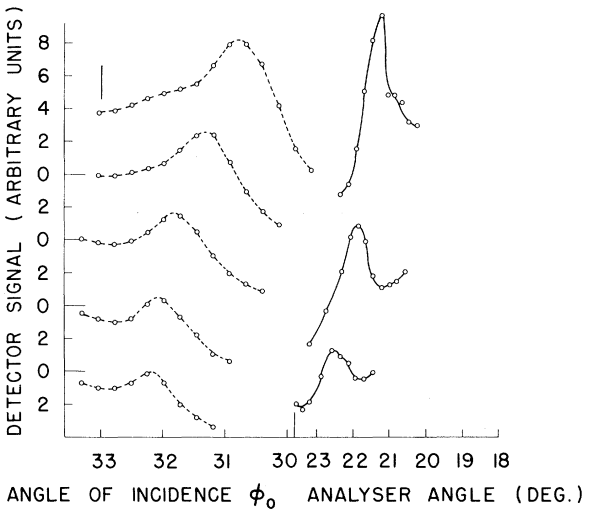

FIG. 3. Intensity maxima observed by scanning in two different modes: (1) On the LHS, the energy transfer ΔE was kept constant while φ_0 was varied. The ΔE values from top to bottom were 7.8, 8.3, 8.9, and 9.3 meV; (2) on the RHS, maxima were found for scanning the incident angle φ_0 and the analyzer setting (k) while maintaining ΔQ constant. The values of $\Delta Q/\Delta Q_{max}$ from top to bottom are 0.25, 0.30, and 0.33. $\Delta Q_{max} = \pi/a$, $a = 1.805$ Å.

(RHS) of Fig. 2 resulted from the elastic component which was observed over a wide range of φ_0 values, i.e., $\varphi_0 \lesssim 25°$. The origin of this component [$\sim 10^{-4}$ of (00) value] is not clear and was observed in our previous work[4a] as well as for Ag(001) also prepared by chemical polishing.[4b] This peak could have arisen from an elastic incoherent component resulting from surface irregularities (point defects, irregular steps, etc.). If this were so, it was indicative of the imperfection of the crystal surface although seemingly the condition was not serious enough to prevent the detection of surface waves or the ordering of Xe atoms on the surface (see below).

The details on the left-hand side (LHS) of Fig. 2 resulted from inelastic phonon-creation events (the magnitude of k was varied and the locus was moved through the hatched region of Fig. 1). The vertical lines in Fig. 2 show where events due to Rayleigh waves were expected. The well-defined peak to the right of this region did not seem to be associated with a surface wave while a small maximum to the left seemed to result from a surface wave (two upper scans). A small bulk detail was also seen which was more clearly resolved in the lower scan. It is observed that all the loss events largely appeared within the limits imposed by momentum changes due to surface waves. The larger sharp feature could be followed out to energy transfers ~8 meV before detection became impossible. This feature was associated with points (solid circle) on or to the left of the line labeled T in Fig. 1, which is the transverse wave branch for Cu taken from Svenssen, Brockhouse, and Rowe.[5] This feature was always associated with gain in the parallel momentum of the beam.

In order to investigate the inelastic region which seemed to arise from surface waves, two other types of scans were performed. Conservation of parallel momentum is given by

$$k_0 \cos\varphi_0 - k \cos\varphi_s = \Delta Q. \quad (1)$$

ΔQ is the change of parallel momentum.

If k_0 and k were kept constant, i.e., constant energy transfer ΔE, and φ_0, φ_s varied, maxima were observed which yielded ΔE for an observed ΔQ. Such scans are shown on the LHS of Fig. 3 in the region of parallel-momentum loss. ΔE values up to ~9 meV were detected and these maxima are associated with the square points of

Fig. 1, where it is seen they lie close to the line labeled R obtained from the Rayleigh wave velocity 2.015×10^5 cm/s given by Farnell.[6]

The other type of scan performed was a constant-ΔQ scan. In Eq. (1), if k_0 was maintained constant, φ_0, φ_s, and k could be varied to yield a constant preselected value of ΔQ which resulted in an energy scan (k varied) for constant ΔQ. The results of such scans are shown on the RHS of Fig. 3 for atom-momentum loss. These maxima yielded points (the ones with uncertainties indicated) which lie close to the R branch in Fig. 1. It can be concluded that in the case of Cu(001):

(1) Energy loss events were observed for both parallel-momentum gain and loss. Both bulk and surface phonons were detected.

(2) Some momentum loss events arise from surface waves, but momentum gain events seemed to arise from bulk modes of vibration.

(3) No energy gain events were observed for either subspecular or supraspecular scattering and energy loss only for subspecular scattering.

In contrast to these conclusions, the results for scattering from an adsorbed Xe layer were totally different. Figure 4 shows the results of He scattering from an ordered layer of Xe on Cu(001) annealed slightly after deposition. The film gave rise to a sharp diffraction peak comparable in intensity to the (00) peak when the Cu crystal was tilted and the energy analyzer set at $\Delta E = 0$. This yielded a layer spacing of 4.1 Å. The out-of-plane diffraction peaks were not accessible with this system so that the geometrical arrangement is not certain. Chesters and Pritchard[7] found that in the case of several faces of several metals, Xe developed a hexagonal arrangement at 77°K rotated often with respect to the substrate.

The scans shown in Fig. 4 were analyzer scans for fixed φ_0 and it can be seen they were similar for all values of φ_0 over a wide range, i.e., the peaks did not shift positions as the incident conditions were changed. This was true for both subspecular and supraspecular scattering. It meant that unlike the results of Cu, the values of ΔE are independent of ΔQ. The scattering experiment detects atom motions which are predominantly perpendicular to the plane of the surface. Therefore, the surface potential well will greatly influence this motion[8] unlike motions in the plane of the film which are largely influenced by Xe-Xe forces. At least two explanations might account for these results: (1) That the motion perpendicular is accounted for by uncorrelated Einstein oscillators. (2) That a resonant mode exists between the Xe atoms and the substrate acoustic modes[8] as the mass ratio Xe/Cu ~ 2.

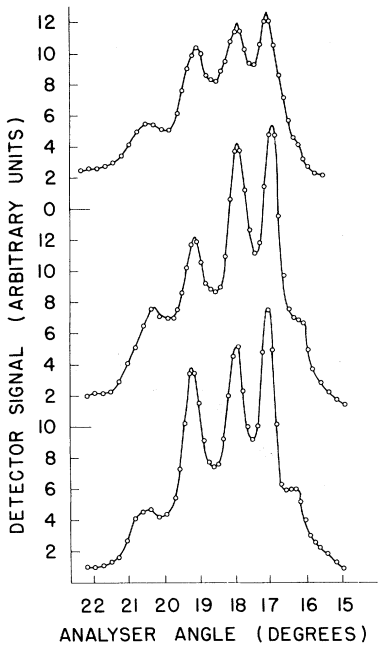

FIG. 4. Scans made by tilting the analyzer crystal for fixed incident angles φ_0 for an ordered arrangement of Xe atoms adsorbed on Cu(001). The values of φ_0 from top to bottom are 16.4°, 29.9°, and 33.0°. Both energy gain and loss peaks are observed which show no variation in position as a function of φ_0. The values of ΔE for the peaks are 2.5 and 5 meV.

The scans in Fig. 4 also indicate that both energy gain and loss peaks were excited, i.e., peaks on either side of the elastic peak at 18°. That energy gain is more likely to occur in the adsorbed system is a function of the lower binding forces than between Cu atoms and the absolute temperature. Energy loss and gain peaks occurred for both subspecular and supraspecular scattering, unlike the case for Cu.

The elastic peak at 18° was still in evidence. Although adsorption might be expected to increase the elastic incoherent component, it was found that progressive adsorption suppressed the elastic component of Cu for all values of φ.

Similar loss and gain peaks to Xe have been found for CO adsorption showing the same general pattern of behavior.

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Dimensionality Crossover in the Organic Superconductor Tetramethyltetraselenafulvalene Hexafluorophosphate [$(TMTSF)_2PF_6$]

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Polarized reflectance measurements from the far infrared to the visible are reported for $(TMTSF)_2PF_6$. At low temperatures, both parallel and perpendicular polarizations show plasma edges in the infrared with metallic reflection below these edges, indicating two- or three-dimensional behavior. The transverse plasma frequency is $\sim\frac{1}{5}$ of the parallel plasma frequency. At higher temperatures, the plasmon in the perpendicular polarization becomes highly overdamped, indicating a crossover to one-dimensional behavior.

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The organic linear-chain compound *bis*-tetramethyltetraselenafulvalene hexafluorophosphate, $(TMTSF)_2PF_6$, is currently of great interest because it remains metallic to low temperatures[1] (19 K) and because it becomes superconducting[2] (when subjected to a hydrostatic pressure of 12 kbar and cooled below 0.9 K). It is the first-known organic superconductor. These properties are somewhat surprising in view of its structure,[3] highly anisotropic dc conductivity, and near-infrared reflectance,[1] which suggest that $(TMTSF)_2PF_6$ is at least as one-dimensional (1D) as tetrathiafulvalene tetracyanoquinodimethane (TTF-TCNQ).[4] It is generally believed, as demonstrated experimentally in the case of TTF-TCNQ, that because of the Peierls transition, quasi-1D systems should exhibit neither superconductivity nor low-temperature metallic behavior.[5]

A recent interpretation based on the pressure dependence of the transition temperatures in $(TMTSF)_2PF_6$ suggests that the material may be electronically 2D or 3D at low temperature.[6] In this Letter we present direct evidence for considerable overlap transverse to the chain direction. With decreasing temperature we have observed the growth of an infrared reflectance edge for electric field perpendicular to the chain direction, which strongly resembles the plasma edge of a Drude metal. Such an edge has not been previously observed in organic linear-chain conductors; we suggest that it results from a significant transverse bandwidth. At high temperature the edge is overdamped. Thus from the optical point of view $(TMTSF)_2PF_6$ must be considered a 1D metal at high temperature, while it is 2D or 3D at low temperature. The crossover from one regime to the other occurs when $kT \sim W_\perp$, where $W_\perp$ is the transverse bandwidth. At higher temperatures thermal disorder localizes electrons on single chains (leading to diffusive trans-