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PROJECT TITLE:

He Atom-Surface Scattering: Surface Dynamics of Insulators, Overlayers and Crystal Growth.

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DISCLAIMER

TABLE OF CONTENTS

I. PROJECT SUMMARY

Investigations in this laboratory have focussed on the surface structure and dynamics of ionic insulators and on epitaxial growth onto alkali halide crystals. In the latter the homoepitaxial growth of NaCl/NaCl(001) and the heteroepitaxial growth of KBr/NaCl(001), NaCl/KBr(001) and KBr/RbCl(001) have been studied by monitoring the specular He scattering as a function of the coverage and by measuring the angular and energy distributions of the scattered He atoms. These data provide information on the surface structure, defect densities, island sizes and surface strain during the layer-by-layer growth. The temperature dependence of these measurements also provides information on the mobilities of the admolecules. He atom scattering (HAS) is unique among surface probes because the low-energy, inert atoms are sensitive only to the electronic structure of the topmost surface layer and are equally applicable to all crystalline materials.

It is proposed for the next year to exploit further the variety of combinations possible with the alkali halides in order to carry out a definitive study of epitaxial growth in the ionic insulators. The work completed so far, including measurements of the Bragg diffraction and surface dispersion at various stages of growth, appears to be exceptionally rich in detail, which is particularly promising for theoretical modeling. In addition, because epitaxial growth conditions over a wide range of lattice mismatches is possible with these materials, size effects in growth processes can be explored in great depth. Further, as some of the alkali halides have the CsCl structure instead of the NaCl structure, we can investigate the effects on the heteroepitaxy with materials having different lattice preferences. Finally, by using co-deposition of different alkali halides, one can investigate the formation and stability of alloys and even alkali halide superlattices.

Other work to be carried out in this laboratory will also be described briefly. This includes experiments with other ionic insulators and a study of the dynamics of the head groups of alkyl thiols which self-assemble on Au (111) and Ag(111). We expect in the case of the alkyl thiols to begin another line of investigations into growth as it is possible to deposit an overlayer which would chemically bond to the alkyl chains. Another potentially interesting system for growth studies with direct technological implications is the natural two-dimensional materials such as the transition metal dichalcogenides.

II. PROJECT

A. Introduction

The scattering of low energy, neutral, non-reactive He atoms from crystalline surfaces is an exceptional way to investigate the static and dynamic properties of surfaces and of epitaxial growth of crystals on the nanoscale.[1-9] The momenta ($4\text{-}14\text{Å}^{-1}$) and energies (10-100meV) of the He atoms match the momenta and energies of the surface phonons, and the atoms do not penetrate the surface. The intensities of the coherently elastically scattered He atoms due to specular and Bragg diffraction provide information on such static properties as surface geometry, corrugation and the He-surface potential.[5,6,9] The inelastic scattering from single phonon annihilation or creation events provides data on the surface phonon momenta and energies from which the surface dispersion curves can be constructed.[1-4,7,8] These characterize the surface dynamics and when the data extend to the Brillouin zone boundary[1-4,7,8] are particularly sensitive to the nature of the forces that reflect the distribution of electronic charge between the surface species, including the effects of strain in epitaxial adlayers.

Other spectroscopies and probes have been used to obtain surface information, particularly electron energy loss spectroscopy (EELS), low energy electron diffraction (LEED) and reflection high energy electron diffraction (RHEED). Both EELS and HAS can be used to measure surface dispersion curves throughout the surface Brillouin zone, but EELS has generally been more useful for the higher energy modes of non-insulators while HAS been better for the low energy modes of all crystal surface materials.[1,10]

The method of choice for the monitoring of growth by molecular beam epitaxy (MBE) has been the RHEED method which has contributed greatly to studies of growth and superlattice preparation in spite of a number of difficulties in interpreting results.[11] HAS, on the other hand, has several advantages over RHEED, particularly for insulators, since the He atoms are neutral, non-penetrating and non-reactive. The elastic scattering studies yield very nice structural features of the growing surface and because of the large adsorbate cross sections leading to the diffuse He scattering, it can also be used to characterize defect densities in real-time.[12-15] Possibly of greater importance, the inelastic scattering of He atoms can provide information on the changing dynamics during the growth, including adsorbate-substrate interactions and adlayer strain which are manifested in the surface dispersion.[16]

Although the proposed investigations depend heavily upon previous He atom scattering (HAS) studies on the structure and dynamics of clean crystal surfaces, this field will only be briefly touched upon below as it has been adequately reviewed in the literature.[1,4,17-25] Instead, the growth studies carried out by our group on the alkali halides will be detailed, which we suggest has set the stage for the proposed work.[12,13]

When He atoms collide with a crystal surface, both elastic and inelastic events take place. The coherent elastic scattering arises from specular reflection and Bragg diffractive scattering.[4-7] The location of the Bragg peaks depends on the dimensions of the unit cell on the surface, and the intensities of the elastic peaks can be related back to the "corrugation" in the atom-surface potential.[5,6]

An additional mechanism for the scattering of He atoms is related to the defects on the surface. This aspect of HAS is only now becoming recognized as a powerful method for observing processes which destroy or modify order on the surface. Defect sites, including isolated adsorbates and terrace steps, give rise to diffuse (<u>i.e.</u>, incoherent) elastic scattering which shows up as loss of intensity in the specular and Bragg peaks, broadening of these coherent peaks, and as large elastic signals at non-Bragg scattering angles in measurements of the inelastic scattering.[4,14,15,26-29]

The inelastic scattering can be considered to result from the interaction of the He atoms with higher-order terms of the interaction potential, which account for the surface phonons and for the bulk phonons projected onto the surface.[1,7] Thus, the inelastic scattering contains the surface dynamics.

B. He Scattering Instrument

The HAS instrument used by our group and to be employed in the proposed work is shown schematically in Figure 1. Briefly, it is composed of several ultra high vacuum (UHV) chambers connected together and has a fixed geometry design: The axis connecting the nozzle beam source and the crystal target makes a 90° angle with the axis connecting the target and the detector. The main features are the nozzle beam source which produces a nearly monoenergetic ($\Delta E/E \approx 2\%$) He beam; the manipulator in the scattering chamber which holds the crystal target; the time-of-flight (TOF) section needed for energy analysis of the pulsed He beam; and the detector, consisting of an electric quadrupole mass spectrometer and a computer-controlled data acquisition system.[2,18]

Each target crystal was cleaved in air and then inserted into the vacuum chamber. The chamber was baked to achieve UHV conditions, and the target was cleaned by sublimation as determined by angular distribution measurements and inelastic studies which could be compared with previous measurements of this same surface. The deposition of the adsorbate was carried out by heating the appropriate second alkali halide crystal so that sublimation could take place from a broad effusive source.[30] A shutter was opened to expose the source to the target crystal (Figure 1). A thickness monitor, not shown in Figure 1, was used to confirm the thickness of the deposited layer.

Three types of measurements were made with this arrangement: 1) intensity vs. deposition thickness for the specular and Bragg peaks, which were carried out under a variety of experimental conditions in real time; 2) angular distributions of the scattered He at different coverages and for various deposition rates, He atom wavevector and surface temperature parameters; and 3) time-of-flight (TOF) measurements at different coverages. The first type of measurement was carried out in real-time, while the latter two were carried out after interrupting the deposition with the shutter. It was found that the growth could be turned on and off repeatably if the substrate temperature was not too high.[12,13]

C. Experimental Results

1. Homoepitaxy-NaCl/NaCl(001)

Figure 2 shows the typical oscillations observed in the elastic scattering intensity with increasing coverage (Θ) for crystal growth of NaCl on itself as monitored by HAS. The factor to convert from time to coverage is on the figure. The total scattering is affected by two principal contributions: 1) Bragg scattering from what remains of the original surface and from the surfaces of the growing island terraces; these can also give rise to interference effects[12,13-15,31]; and 2) diffuse elastic scattering from the steps of the growing terraces and/or from isolated adsorbates. In addition to these, there may be contributions from the changing inelastic background. Complicating the picture further are the dependence of the island growth on the substrate temperature and the deposition rate[11,12,15,31-33] and that the density of defects may build up non-uniformly with the number of layers, possibly arising in part from poor geometry in the source or scattering configuration.[30]

A reasonable explanation, then, for the main features observed in Figure 2 follows directly.[11,15,31,34,35] 1) The initial decrease in the coherent scattering intensity at very small coverages (i.e., $\Theta \leq 0.1$ monolayer) is due primarily to the diffuse scattering from the isolated NaCl adsorbates. 2) Island build-up occurs as Θ approaches approximately 0.5 monolayer so that the Bragg scattering intensity reaches a relative minimum due to the diffuse scattering from the terrace steps and, to an extent depending on the He wavevector, to destructive interference between the coherent scattering from the original surface and from the large terrace areas formed from coalescing islands. 3) The intensity rise from $\Theta=0.5$ to $\Theta=1.0$ is due to the decrease in step scattering as the terraces continue to grow together. 4) The overall decrease in intensity with number of layers results from the starting of new islands before the previous layer is completed and the concomitant build up of a constant defect density under these growth conditions.

Although complicated by a number of contributing effects, most of the basic features noted above have been interpreted within the framework of the kinematical approximation, just as for electron scattering via RHEED and LEED.[1,11,14,15,31-34,36-38]

To characterize the deposition we have employed a number of different procedures. The simplest is to stop the deposition at the desired coverage and measure the angular distribution. Examples of this are shown in Figure 3 where the symbols label the Bragg peaks. One can calculate the surface lattice spacing from the angles of the peak positions and the initial He wavevector, k_i, using the Bragg scattering relation between the surface reciprocal lattice vector and the momentum transfer parallel to the surface, $\mathbf{G} = \Delta\mathbf{K}$, which for this instrument (See Figure 1) is given by[18]

$$\Delta K = k_f \sin\theta_f - k_i \sin\theta_i = \sqrt{2} k_i \cos(\theta_i + 45°) \qquad (1)$$

Since the surface lattice spacing $a_s = 2\pi/G_{10}$, one obtains $a_s = 3.99$Å for NaCl, in good agreement with that calculated from the bulk fcc lattice spacing.[39] From the FWHM of the specular peak of the clean surface ($\sim 0.1°$) one can estimate the transfer width of the instrument to be about 550Å for this value of k_i.[15]

Even though the specular and Bragg peak positions remain the same throughout the deposition, the peak intensities, the background and, most importantly, the peak profiles change dramatically as a function of incident wavevector, as predicted by the kinematical approximation.[40,41] These profile changes are quite evident in Figure 4 which presents measurements for $\Theta = 0.5$ of three full angular distributions and their respective first-order Bragg peaks displayed on an expanded angle scale. Note that we have converted the abscissa variable from angle to surface momentum exchange ΔK. This is a more useful presentation here because the Bragg peaks remain at the same values of ΔK for different incident wavevectors.

Figure 4 shows that the intensity of the Bragg peaks appears to be made up of two approximately Gaussian functions whose amplitudes vary with incident wavevector.[40,41] If the crystal growth is predominantly two-dimensional, then interference can arise between the coherently scattered beams from the new growing terraces and the exposed portion of the old surface observed at a Bragg angle θ_i (Equation (1)). This gives rise to a "phase condition" for the scattered waves, which depends on the perpendicular or z component of the wavevector,[15,31,33,34,36,40] $|\Delta\mathbf{k}_z| = |\mathbf{k}_{fz} - \mathbf{k}_{iz}| = N(2\pi/D)$. Again, for this HAS instrument

$$|\Delta\mathbf{k}_z| = \sqrt{2}k_i\sin(\theta_i + 45°) = N(2\pi/D) \qquad (2)$$

where the subscripts f and i refer respectively to the outgoing and incident wavevectors, D is the step height and N = n, an integer, for constructive interference and = n+½ for destructive interference. The kinematical approximation thus predicts an oscillatory behavior for the profile of a particular Bragg peak with k_i.

Figure 5 shows a plot of the ratio of the maximum amplitude of the broad peak, A, to that of the sharp one, A_0, as a function of $N = \Delta k_z D/2\pi$. The value of D can be fit experimentally to the above expectations for Bragg and anti-Bragg scattering conditions. We find D = 2.8Å, or one-half the bulk unit cell length ($a_b/2$), as expected for NaCl.[39] The dashed curve is the expected ratio for one-half monolayer coverage, slightly modified from the kinematical approximation.[41]

Also in Figure 5, the solid set of points are the similar ratios taken for the surface prior to the deposition. These data do not show any oscillatory pattern; hence, clean cleaved NaCl appears to be a very good surface indeed! This is consistent with electron micrographs of NaCl surfaces decorated with gold atoms that show terrace widths of a few thousand Ångstroms[42], several times the instrument transfer width.[14,15]

Under the conditions of very low surface coverage, the reduction in the specular and Bragg beam intensities by the adsorbates can be treated in an analogous manner to gas phase scattering.[15] Thus, one can take the intensity of a coherently scattered peak to be given by

$$I_G = I_G^o\exp\text{-}(\Sigma\Theta n_s) \approx I_G^o(1 - \Sigma\Theta n_s) \qquad (3)$$

as $\Theta \rightarrow 0$. Here Σ is the surface cross section for one adsorbate unit, Θ again the fractional coverage, n_s the density of surface sites and I_G^o and I_G refer, respectively, to the specular or Bragg intensity initially and after deposition.[15] The Σ values which we determine from the initial slopes in the deposition curves reflect a composite of all processes which deplete the coherent scattering channels in the limit of very low coverage.[15,43]

In Figure 6, the cross section measurements are plotted as a function of inverse target temperature for k_i about 7.7Å^{-1}. At temperatures up until about 250K the cross sections appear to be relatively flat, consistent with isolated molecules, and then at higher temperatures they drop substantially. This "transition" temperature is somewhat higher than expected as Yang and Flynn have reported[44] that alkali halides adsorbed on alkali halides possess considerable mobility at temperatures above about one-ninth of the melting temperature, for NaCl about 110K. Our picture is that above 250K the relatively isolated adsorbates nucleate as islands, which has the effect of presenting smaller cross sections for the same nominal coverage. An analogous decrease is noted for epitaxial growth of metals on metals[14] except that instead of reaching a step, a NaCl is more likely to encounter another molecule.

Engel and co-workers[14] have proposed a simple model to estimate the size of the islands from the dependence of the specular beam intensity on Θ for "in-phase" scattering conditions. According to Equation (2) when N is an integer, the decrease in specular intensity should come predominantly from the diffuse scattering due to the presence of island steps. These workers obtain an expression for the intensity which is equivalent to

$$I_0/I_0^\circ = 1 - (2^{1/2}/r_o)(\Sigma\Theta^{1/2}) \qquad (4)$$

where r_o is the effective island radius at $\Theta = 0.5$ and Σ here is the cross section per unit step length. (This formula should not apply in the very low coverage regime with adsorbates very far apart.) Figure 7 shows a plot of the specular intensity vs. $\Theta^{1/2}$ for data taken at 210K, 318K and 375K. The slopes of the curves give values for r_o as 15Å, 43Å and 74Å, respectively. Perhaps more important than these model-dependent values is the confirmation that the island size appears considerably greater at the higher temperature, which is consistent with the cross section measurements above.

All in all, the results of the homoepitaxial growth of NaCl onto NaCl are not so different from what one would expect given the temperature dependence of the admolecule mobility and the results of similar experiments on metals and semiconductors. However, the results for heteroepitaxy, described next, does provide new insights into growth behavior.

2. Heteroepitaxy-KBr/NaCl(001)

Figure 8 shows the behavior of the specular signal during deposition of KBr on NaCl (001) which have a lattice mismatch of about 17% and reveals some very interesting features.[13] First, one can see that the drop-off is very steep. At low coverages we find values for Σ of about 200-250Å^2 at 223K which is in the range found for species like isolated CO molecules on metals[15], but which is considerably larger than has been measured for NaCl deposited on NaCl (~80-100Å^2) as shown in Figure 6.[12] One should note that the large cross section for CO may be due in part to its orientation normal to the metal surface.

Second, by measuring the times for the deposition of one monolayer (the peak-to-peak differences), one sees that from the beginning of the deposition to the position of the first peak a period of time corresponding to approximately 2 monolayers has elapsed. Thus, it appears that the first monolayer deposited does not form a "good" surface which can lead to coherent specular intensity. In the homoepitaxy of NaCl, the first oscillation appears at the expected time, Fig. 2.[12] This "missing" oscillation behavior has also been found in a few other epitaxial studies.[45,46] For the case of NaCl deposited onto Ge[46], an explanation has been offered that the NaCl deposits initially in an orientation normal to the surface, in effect for these ionic molecules, building up a double layer; the following layers would then be expected to grow by adding molecules with a parallel orientation. In view of the fact that the cross section for KBr is comparable with that of CO and is much larger than that of NaCl, which probably deposits parallel to the NaCl surface, a similar initial growth mechanism might apply here.

In Figure 9 are shown the angular distributions corresponding to the positions labeled in Figure 8; that is, angular distributions for the clean NaCl surface and for each additional monolayer from 2 through 6. The diffraction peaks are still given by the Bragg scattering condition of Equation 1. In the diffraction for 2 monolayers, the peaks for NaCl are still very evident as are the small "superstructure" peaks which correspond to a corrugation length of about 28Å (for the surface unit mesh in the < 110> direction, or $28\sqrt{2}$Å in the < 100> direction) equivalent to the dimensions of 7 NaCl's or 6 KBr's (based on the lattice spacings of each crystal, 3.99Å for NaCl and 4.66Å for KBr).[39] Much smaller here are the peaks corresponding to the KBr lattice dimensions. As the number of layers increases, the NaCl Bragg peaks diminish in intensity and the KBr Bragg peaks grow in. The superstructure peaks at the same time at first increase and then, by 6 monolayers, disappear. It is very likely that the superstructure corrugation corresponds to the "buckling" of the deposited layers under the strain of the lattice mismatch.

Not shown in Figure 9 are angular distributions measured in the < 110> direction for the same coverages. These show the 28Å superstructure behavior in just the same way. Further, in additional work in this lab on the deposition of NaCl onto KBr (001), the complementary system, we find very similar behavior with the NaCl peaks growing in and the KBr peaks fading out over a period of about 7 monolayers; and during this period a similar 28Å corrugation is in evidence.

To test whether the complex patterns shown in Figure 9 can arise just from the superposition of the waves corresponding to the nominal corrugations of NaCl and KBr, we have begun to carry out simple eikonal calculations.[47] This kind of approach to modeling the diffraction may seem reasonable for the alkali halides because the forces at work are predominantly Coulombic and very similar for both adsorbate and substrate; the buckling is due to an accommodation of the different ion sizes and a competition between the intermolecular potentials of like and unlike molecules.[44,48-51] One might expect that the situation would be far less simple for metals and semiconductors, or for molecular solids, where the electronic configurations of the substrate and adsorbate need to be dealt with. However, the size of the electronic cores of the interfacing species may still play the major role in determining the <u>shape</u> of the interface. This lattice matching situation has been recognized previously and is given by the Vermier-Nonius relation.[48] However, it may be a general rule that for species A and B with surface lattices a_A and a_B, the lattice mismatch will lead to a corrugation for the buckling with length $L = n_A a_A \approx n_B a_B$, where the n's are the smallest whole numbers satisfying the near equality, because this leads to a minimum in the surface free energy.

Preliminary time-of-flight (TOF) measurements for the inelastically scattered He atoms have been carried out at a coverage of 4 monolayers. Figure 10 shows a series of spectra, taken at different non-Bragg incident angles, θ_i, to illustrate the quality of the data. The first point to notice is that the largest peak in each spectrum is the diffuse elastic peak, labeled E. This peak is a sensitive measure of the surface quality since it arises from defect scattering. Its prominence in these spectra stands in sharp contrast with the rather small diffuse elastic peaks found in the TOF spectra from cleaved KBr.[18] Secondly, several sharp peaks in each spectrum, corresponding to the creation or annihilation of single phonons, rise above the background. From the positions of the peaks relative to the elastic times, path length (1.04m) and incident and scattering angles ($\theta_f = 90° - \theta_i$), the phonon frequency ω and surface wavevector **Q** can be calculated from the "conservation laws",

$$\hbar\omega = (k_f^2 - k_i^2)\hbar^2/2m \qquad (5)$$

and

$$\Delta\mathbf{K} = \mathbf{G} + \mathbf{Q} \qquad (6)$$

and, hence one can obtain the surface phonon dispersion $\omega(\mathbf{Q})$.

The peaks in Figure 10 have been labeled according to their position near a surface-localized mode or resonance which had been previously calculated.[52,53] In our identification scheme for Figure 10: R stands for the Rayleigh mode, the lowest energy sagittal plane acoustic mode, designated S_1 by de Wette and co-workers[52,53]; L for the acoustic longitudinal resonance or S_6; C for the crossing resonance which is the surface projection of sagittally-polarized, transverse acoustic bulk modes at the boundaries of the bulk Brillouin zone, sometimes designated S_8[22]; and S_2 for the sagittal plane optical mode which originates with the longitudinal optical bulk mode as $Q \rightarrow 0$. For KBr the S_2 mode lies in the gap between the acoustic and optical bulk bands and is particularly strongly seen in HAS.

The TOF peaks have been converted to dispersion curve points via Equations (5) and (6) and are plotted as the dark squares in Figure 11. The open circles (and the few open triangles) are data obtained previously for a clean freshly-cleaved surface of KBr[18]; the shaded regions are the surface projections of bulk bands and the lines are surface localized modes from the Green's function calculation by Benedek and Miglio.[54]

In general the agreement between the 4 KBr monolayer data and the cleaved KBr crystal data is remarkably good. Aside from the good agreement, four features should be noted: 1) The surface dispersion curves for NaCl lie considerably higher than those for KBr and there is no evidence for them here.[21,24] HAS is sensitive to the topmost layer only. 2) The data for the Rayleigh mode do not go to zero at the $\bar{\Gamma}$ point, but rather to a finite value. This is to be expected for a small number of layers whose vibrational frequencies are not perfectly matched to those of the substrate.[55-57] 3) We have very few data points for the longitudinal (S_6) mode. Longitudinal vibrations are more sensitive to surface imperfections than the Rayleigh wave is. Hence, for a surface with many small islands the longitudinally polarized phonons have short lifetimes and show up as relatively weak TOF peaks. 4) The S_2 data seem to bulge upwards somewhat from the previous data around the middle region of the surface Brillouin zone. It is generally expected that the optical modes should be more sensitive to surface strain than the acoustic modes and this could very well be its microscopic signature.[16]

The angular distribution in Figure 9 for the 4 monolayer surface shows both KBr and superstructure diffraction peaks. Thus, the "true" Brillouin zone for this surface corresponds to the 28Å lattice, and the data shown in Figure 11 should be folded 6 times. That is, the $Qa/2\pi$ values of 1/6, 3/6 and 5/6 lie at the $\bar{M}$ point, while 0, 1/3, 2/3 and 1 lie at the $\bar{\Gamma}$ point. One might expect to see splittings at the "true" zone boundaries, or folding points, but our data are not good enough to distinguish these.

In Figure 11 two data points appear below the Rayleigh wave. As they do not occur in spectra taken near Bragg peaks, they cannot be due to deceptons.[2] It is possible they are due to the folding mentioned above, but this will need to be examined further.

A qualitative picture of the heteroepitaxy of KBr onto NaCl (001) comes primarily from the deposition curves and angular distributions at this stage. The diffraction pattern in the angular distribution, after the deposition of two monolayers, shows peaks with both the NaCl and superstructure corrugation, and very small specular intensity. This indicates that the initial deposition of KBr must be largely pseudomorphic with the NaCl lattice, but with many defects or very small islands. One possibility, given the lattice mismatch, is for the KBr initially to deposit oriented normal to the surface forming small bilayer patches that buckle and give rise to the superstructure. This arrangement would also have a considerable amount of three-dimensional stacking.

At 3 monolayers, Figure 9, the superstructure peaks are distinctly larger and also sharper in angular width, the KBr peaks are growing in a bit and the NaCl peaks are declining with the NaCl second order Bragg peaks barely visible (and unmarked in the figure). The specular peak has decreased to its lowest value as an oscillation maximum in the deposition curve. Thus, it appears that the patches have begun to smooth out and fill in so that the NaCl corrugation is reduced and the KBr corrugation shows up more strongly; but defects still abound and island sizes remain small. By 4 monolayers the surface quality has greatly improved as the specular and KBr peaks, now including second order KBr peaks, have grown substantially; the NaCl corrugation is no longer observable. By 6 monolayers the specular is much larger indicating a considerably better surface and the superstructure peaks are only barely visible. However, the Bragg peak widths suggest that the size of the terraces is still significantly smaller than the transfer width of the instrument (from the initial specular beam width for $k_i = 7.4Å^{-1}$, about 700Å[15]) in contrast to the broad terraces of cleaved alkali halide surfaces.[42]

3. Heteroepitaxy- KBr/RbCl (001)

Deposition curves (Figure 12) for the KBr/RbCl(001) system, where the lattice mismatch is less than 0.5%, give an oscillatory behavior as in NaCl/NaCl (001) with an important exception: The first and second oscillations do not vary together as the He wavevector is changed, but rather seem to be out-of-phase with each other. This can be seen in the dependence on the incident wavevector. This is interpreted as due to a different step height between the first layer (KBr on RbCl) and the second layer (KBr on KBr). In addition, there is no indication of superstructure in the angular distributions. Thus, the preliminary analysis suggests that because of the near lattice match there is no buckling of the adlayer, but the different sizes of the individual ad-ions alter the surface corrugation in the first ad-layer and the He-surface potential.

Additional information on the growth of the first and subsequent layers lies in the intensity of the first order Bragg peak as a function of coverage Θ as shown in Fig. 13. When the flag is opened, the Bragg intensity initially decreases but then rises very steeply. We believe the explanation for this lies in the fact that even though the lattice mismatch is small and pseudomorphic growth is taking place, the ionic radii of the K^+ and Br^- are different than the radii of Rb^+ and Cl^-. The net result is a large corrugation in the first KBr adlayer. However, in subsequent layers the corrugation returns to that of KBr and the further growth looks much more like homoepitaxy.

Work is still in progress on the energy analysis experiments for this system. However, we have remeasured the surface dispersion curves of the clean RbCl crystal, repeating the first experiments done with this HAS instrument.[17] We found that our resolution has been improved greatly in the last three years so that we now can observe the gap surface optical mode, S_2, similar to the one we reported for KBr.[18] In preliminary experiments with a monolayer of KBr on RbCl the energies of this optical mode seems to be shifted to lower values.

4. Interpretation of the Growth Studies

Up through this stage of the investigation the interpretation of the experiments has been qualitative for the most part. In recent months several theoretical groups have become intrigued by these results and have discussed developing a collaboration. Two of these groups, Professor Celli at the University of Virginia and Professor Manson at Clemson University, have worked with us previously. In addition, Professor R. B. Gerber of the Hebrew University in Israel has become interested in some aspects of the scattering problem.

D. Proposed Experiments

The crystal growth experiments at FSU have been focussed on the alkali halides because they are the prototypical ionic insulator materials. The unexpected richness of this work, which has been described rather extensively above, presents the opportunity for a definitive study into insulator crystal growth. We thus plan to build from these studies along two broad paths: First is to probe further into the epitaxial growth of ionic insulators by fully exploiting the variables available within the alkali halide family and then extending the experiments to include other ionic insulators. Second, the recent advances with self-assembling alkyl thiol films on Au(111) surfaces appears to offer a marvelous opportunity to study chemisorption and growth on insulators. To investigate these surfaces, which are more intricate than alkali halide crystals but are correspondingly more important technologically, will require further development of He atom scattering techniques and also theoretical modeling. Finally, we discuss opportunities with growth on natural layered materials, such as transition metal dichalcogenides, which offer basic and technological interests.

1. Epitaxial Growth on Alkali Halides.

Studies using RHEED as a monitor for the real-time growth have been reported on alkali halides[44,49]. The authors advance the argument that ionically bonded crystals grow under different conditions than metals or semiconductors. We believe our results above demonstrate that HAS should be the method of choice for an investigation on ionic materials and, further, that many features that we have found have also been observed in other systems.

The extension we propose for the alkali halide systems, in addition to the on-going experiments on KBr/RbCl, include 1) additional variations with lattice mismatching, 2) growth on alkali fluorides, 3) growth of materials that normally prefer other lattice structures and possibly 4) the formation of alkali halide alloys through co-deposition. As in the work described above, we plan in each system to characterize the growth by measuring deposition curves, cross sections, angular distributions and the surface dynamics as a function of temperature, growth rate, incident wavevector and co-deposition combinations. So far we have studied KBr/NaCl with a lattice mismatch of 17% and KBr/RBCl with a lattice mismatch of less than 1%. Under 1) we would consider RbI/NaCl(001) which provides a large mismatch of over 30% and NaBr/NaCl(001) for a moderately small mismatch at about 6%. These systems do not need to be carried out in as great a detail as the systems that have already been studied, but it is important to verify the trends.

Yang and Flynn[44,49] comment that heterolayers among the alkali halides can be readily prepared except for the alkali fluorides which do not grow very well. Hence, in 2), we would try similar growth studies with systems like NaCl/LiF(001) and CsF/NaCl(001) to see if we can characterize the differences between the fluorides and the other halides. Using the fluorides also allows us a wider range in lattice matching conditions.

For 3), we are fortunate that the cesium halides except for CsF have the simple cubic CsCl structure. Thus, it will be possible to explore the conditions for growing an adlayer with the "wrong" lattice. We can choose the extent of lattice matching or mismatching to enhance the effects. For example, we estimate that for fcc growth CsCl/KI(001) would have a mismatch of only 2% whereas CsI/KI(001) would be about 12%. We would expect, perhaps naively, an initial strained pseudomorphic growth that would switch over to simple cubic as the thickness increased. What happens in between these two growth regimes could be very interesting for the fundamental science of growth.

Co-deposition experiments in 4) may prove feasible to determine the characteristics of metastable disordered alkali halide alloys such as KCl_xI_{1-x} which exist in the bulk phase diagram. If so, this could be an analog to semiconducting materials like $GaAs_xSb_{1-x}$. Also, from our initial work we believe that the techniques are well enough developed to build superlattice structures of the alkali halides which have interesting structural features. At the present time theoretical modeling of the superlattice behavior of the alkali halides has not been carried out. However, this work has been done for metal combinations and semiconductor superlattices by a number of authors including our collaborators from the University of Milan. We anticipate that a postdoctoral associate from Milan will visit our laboratory to work on such theoretical problems. (See Section VI of this Report.)

We have developed a new device for x-ray alignment and subsequent crystal polishing. It appears to be reliable to 0.05° which is a substantial improvement over current goniometers which are good to 0.25°. The alkali halides form rather good flat surfaces, with steps averaging typically more than 1000Å apart. If this device works as preliminary results indicate, we could prepare vicinal surfaces reliably with a wide variety of step spacings (perhaps sufficiently suitable to act as templates for quantum wires). This would add another important dimension to the growth characterization experiments.

From our past experience we recognize that the experiments outlined above constitute a large undertaking which are not likely to be completed in a year's time. We expect, however, to complete 1) and 2) and a large part of 3) this year, with perhaps some preliminary attempts at 4).

2. Additional Experiments in this Laboratory.

In a proposal submitted recently to the NSF we have requested support for two students, in addition to the three included in the budget request here, who would work on the following systems: (a) other ionic insulators and (b) alkyl thiol films on Au(111).

(a) The experiments on the ionic insulators MgO, NiO and $KTaO_3$ require only a slight modification from the approach used with the alkali halides; namely, these materials need to be cleaved in vacuum. We have constructed a cleaver for in situ cleaving just for this purpose. The initial work on these materials would begin in the Prep Chamber where LEED and AES (Auger Electron Spectroscopy) work could be carried out. When the HAS instrument is free, then the prepared crystals could be transferred under vacuum to it. We would first measure the surface dynamics of the clean crystals since only for MgO has there been any extensive surface dynamics studies.[58-63]

For MgO we plan several types of growth studies: First, the homoepitaxy of MgO; then the deposition of NiO which has less than a 1% lattice mismatch with MgO;[39] and finally, the deposition of Ca for which it has been predicted that a half monolayer coverage should be stable.[58] It should be relatively easy to carry out the last of these depositions in the HAS instrument with a Knudsen source. For the first two we will attempt to produce the oxides in the HAS instrument using the Knudsen source for the metals while co-producing O atoms from an RF discharge source. If this does not work out, we will be able to collaborate with a group at FSU that has developed an MBE laboratory for epitaxial growth of magnetic superlattices.[64,65] The NiO studies will be similar to those suggested for MgO.[66-69]

The $Ktao_3$ crystal has a lattice spacing of 3.99Å. Thus, the K and O surface has very nearly the same geometry and size as that of the NaCl(001) surface. Therefore, after the initial experiments on the clean surface have been carried out, the deposition of NaCl (and possibly other alkali halides) seems like the most rational starting point for further epitaxial growth experiments. These will be followed with the deposition of NiO which has a lattice mismatch of just under 5%.

(b) Chemisorption of hydrocarbon chains onto metal surfaces involves the formation of an interface and a new layer whose order depends on the ordering of the substrate and the interaction with the substrate. Since the hydrocarbons are insulators, the probe of choice with nanoscale resolution would seem to be the neutral He scattering.

Vogel and Wöll have reported on the structure and dynamics of fatty acid monolayers deposited on noble metal crystals, using LEED and He atom scattering.[70] Their LEED results indicate that the Langmuir-Blodgett films form ordered layers on the Cu, Ag and Au (111) surfaces; however, these studies have been difficult to repeat.

Recent publications have reported studies by EELS, XPS (x-ray photoelectron spectroscopy), TPD (temperature programmed desorption), AES, and LEED which characterize monolayers of adsorbates of the general form $HS(CH_2)_nX$ or $(S(CH_2)_nX)_2$ on single crystal Au(111) surfaces, where $X = CH_3$, CO_2CH_3, CH_2OH, CO_2H and $CONH_2$, and n ranges up to 21.[71-83] The results show that an ordered layer is formed, which has each S bonded to a gold atom. The attached hydrocarbon chains sticks upward with a range of angles from 28° to 40° with the surface normal in an all trans configuration. Elastic He atom scattering experiments have also been carried out on these systems.[84] The diffraction patterns obtained show that for $CH_3(CH_2)_{21}S$ the methyl groups form a nicely ordered surface with hexagonal structure and a lattice spacing of 5.01Å. For these samples the domain sizes appeared to be $\leq$ 100Å. It should be noted that the He atoms are probing the top group of the chain and not the S-Au region at the metal surface.

These systems are of interest in general as self-assembling systems and for their potential through the choice of X groups for chemical synthesis. One can imagine a number of uses for an ordered hydrocarbon (or substituted hydrocarbon) layer, including the possibility of designing very specifically binding catalysts, as models for biological interfaces and membranes and for their relevance to corrosion protection and adhesion problems. Indeed, electron transfer processes in the substituted alkyl thiols/gold surface system have been studied as a model system for long-range electron transfer which may occur in cells as for example in photosynthesis.[85]

This work will begin by preparing and characterizing a Au (111) single crystal, first by LEED in a Preparation Chamber and then by HAS to reproduce the literature results.[86] Then, the self-assembling systems will be prepared on the gold surface and similarly characterized. Because of reported differences in the behavior of the alkyl thiols on silver,[87] we would like to repeat the experiments on the Ag (111) surface. We plan to explore the variation in these properties with the different X groups listed above and with temperature. Finally, it would be exciting to deposit a layer of metal, such as Cu, or other material on top of the alkyl chains, which would chemical bond via the X group to them. This could be accomplished either with the MBE instrument or possibly with the new MOCVD apparatus which has recently come on-line in the Chemistry Department. Elastic He scattering experiments have already been carried out on physisorbed layers of Xe, Kr, H_2O, CH_3OH and n-hexane on $CH_3(CH_2)_{21}SH$/Au/mica.[88] It would be most fascinating to apply the HAS techniques to a chemisorbed overlayer system.

3. Future Experiments on Growth.

Two-dimensional layered materials, including the transition metal dichalcogenides, monochalcogenides and tin sulfide and selenide, also can be made to grow epitaxially.[89-92] Because the van der Waals interlayer forces are weak and the covalent intralayer forces are strong, the latter dominate the growth. The interface between layers can tolerate a large misfit which appears to be similar in some ways to that observed in the KBr/NaCl system; namely, it displays a superstructure which depends on the mismatch of the unit cells.

These materials have interesting and potentially useful electronic properties which can be varied from insulating to superconducting by changing the metal or the chalcogen. Further, intercalation compounds can be formed which have modified electronic properties. For example, depositing Cu on SnS_2 leads to diffusion of the Cu into the bulk which converts the SnS_2 semiconductor from a n-type to a p-type. On the other hand deposited Ag reacts with the surface while Au just forms a surface adlayer.[90]

One of us (JGS) has already had considerable experience with these materials, having carried out HAS studies on 2H-$TaSe_2$, 1T-TaS_2 and GaSe at the Max Planck Institute in Göttingen, Germany, and is quite familiar with preparing clean surfaces of them.

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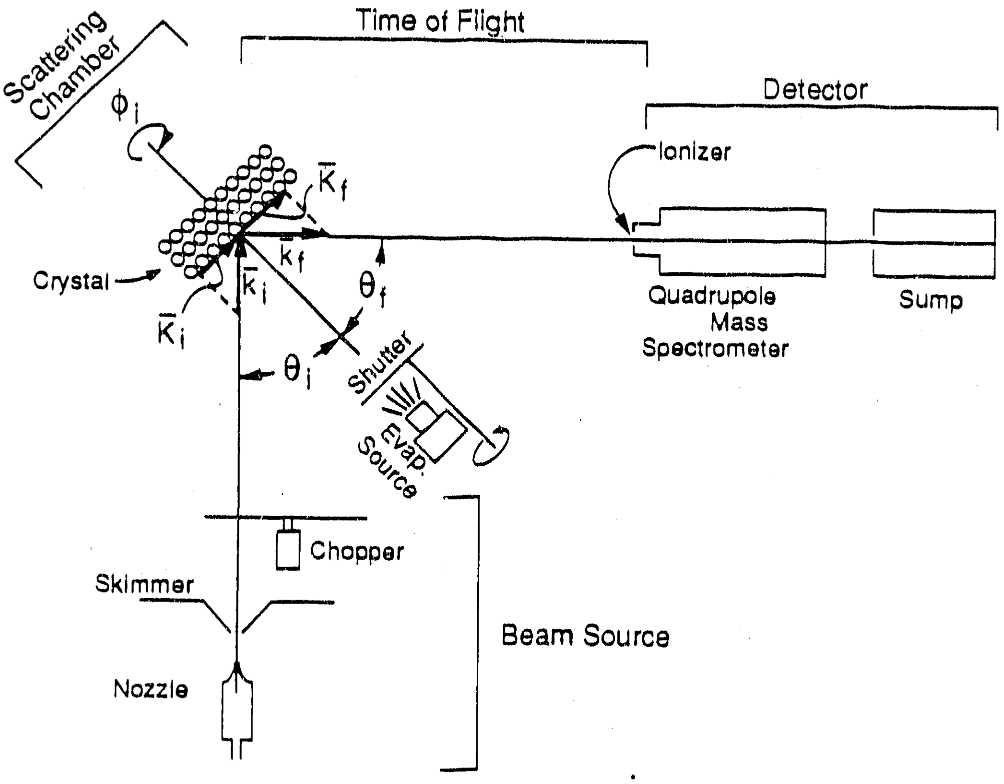


Figure 1. Schematic view of the HAS instrument.

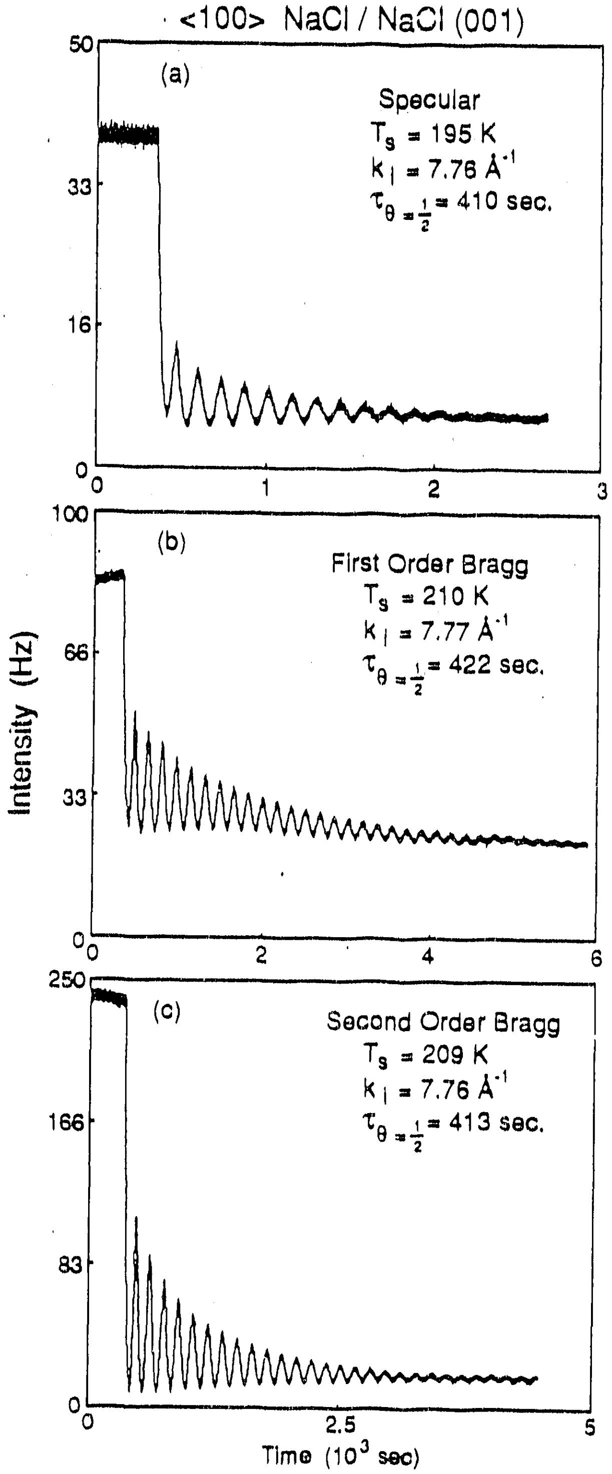


Figure 2. Plots of scattered intensity **vs** deposition time (coverage) for He scattering from a NaCl(001) crystal surface for the specular and the first and second order Bragg peaks. The target temperature, T_S, the incident wavevector, k_i and the time $\tau_{\theta=½}$ to deposit the first one-half monolayer are given in each of the panels. The coverage rate was confirmed by a thickness monitor.

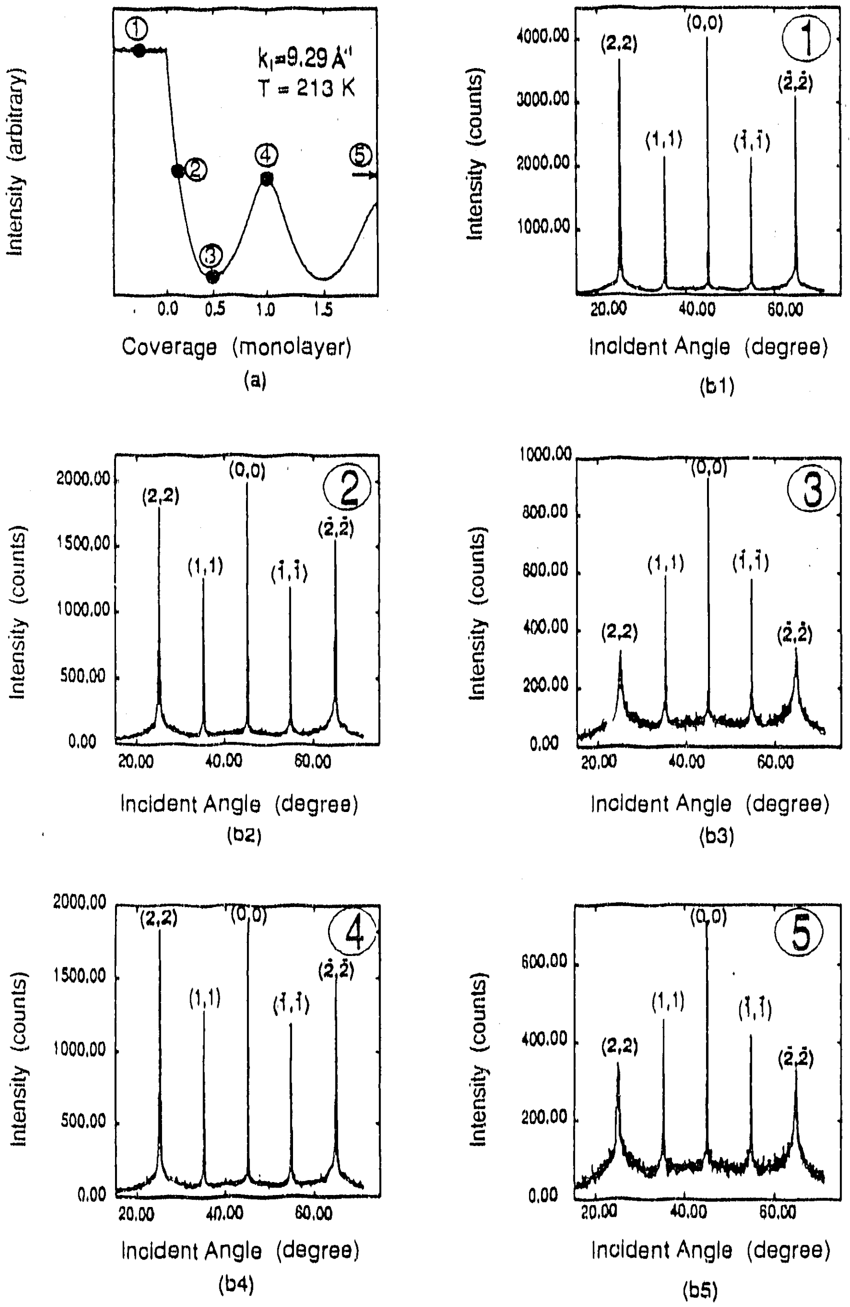
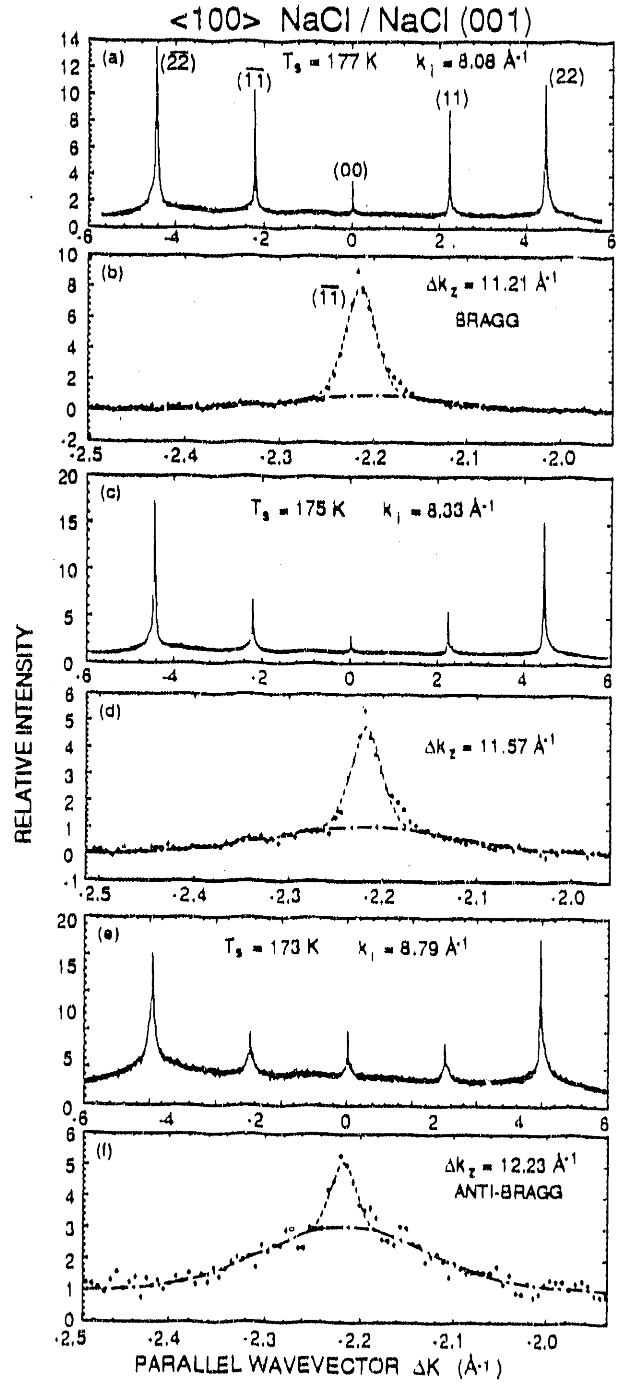


Figure 3. Panel (a) shows a schematic view of the specular intensity vs coverage, where the circled numbers indicate the coverages at which angular distributions were taken; they are shown in (b1) through (b5).



<u>Figure 4</u>. Plots of angular distributions for one-half monolayer coverage for several different incident wavevectors. The Bragg peaks have been labeled. Panels (a) and (b) have a wavevector corresponding to the Bragg condition (constructive interference) and panels (e) and (f) are for the anti-Bragg condition (distructive interference). Panels (c) and (d) are for a wavevector condition in-between. Panels (a), (c) and (e) are the full angular distributions and panels (b), (d) and (f) are the first order Bragg peaks on an expanded scale.

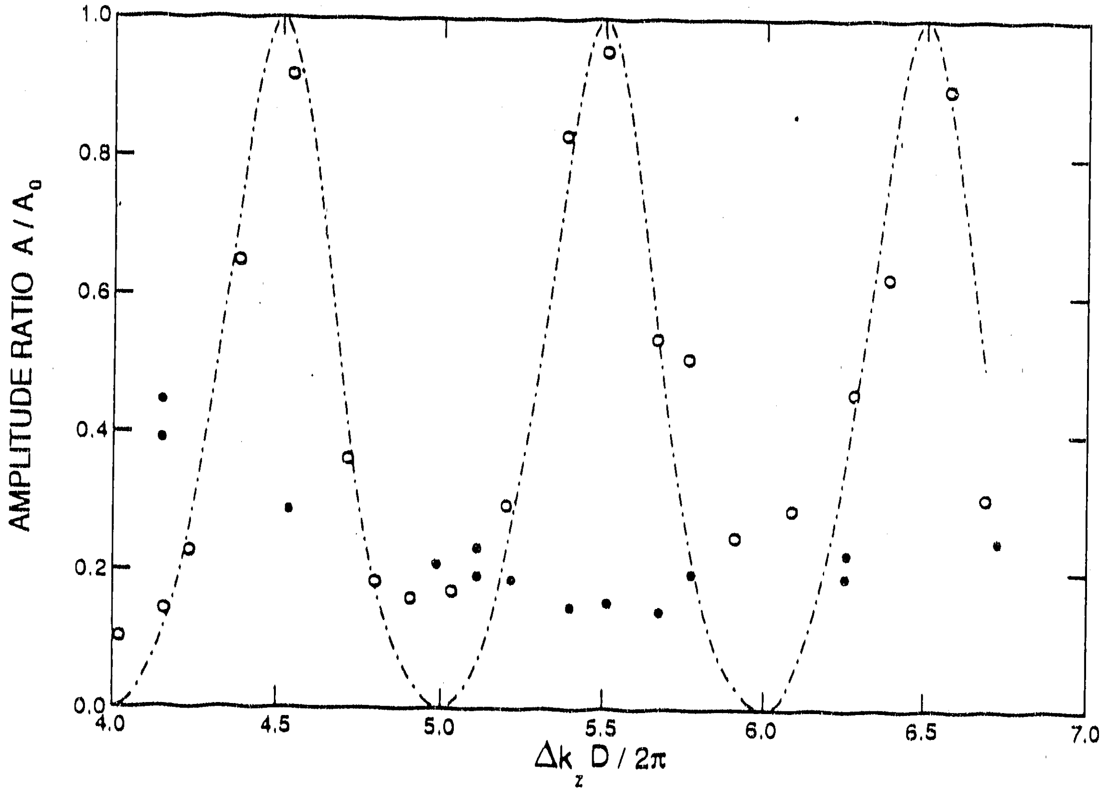


Figure 5. Plot of the amplitude ratios from results as in Figure 4 vs $N = \Delta k_z D/2\pi$, as described in the text. The open points are for one-half monolayer coverage. The solid points refer to the amplitude ratios for the initial clean surface.

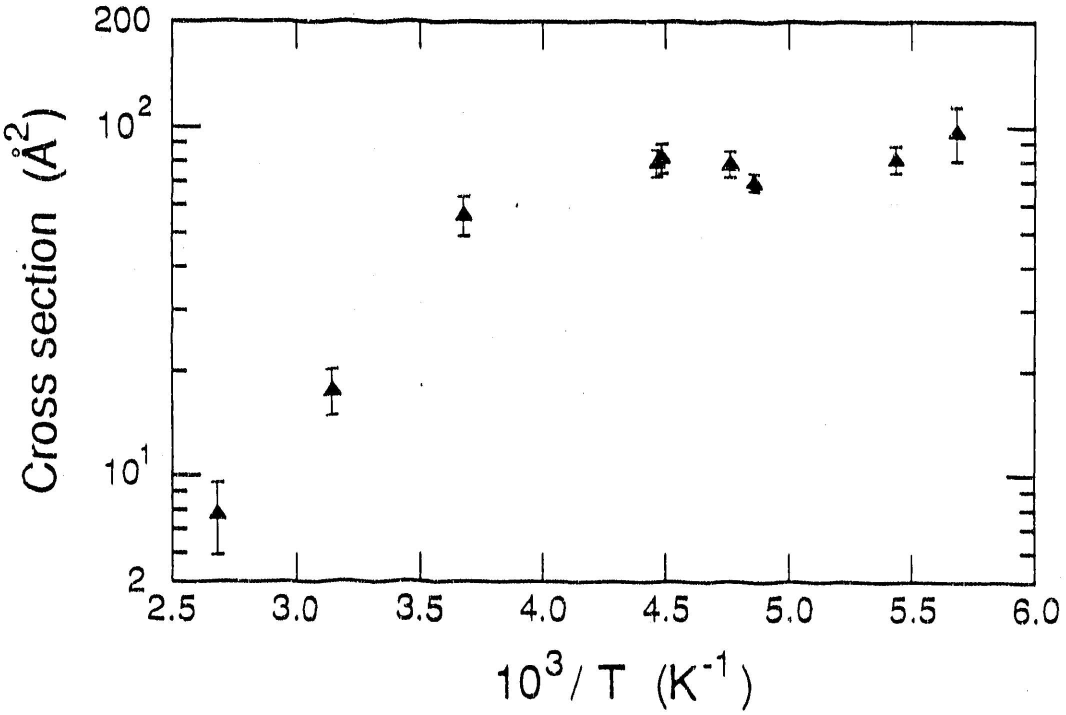


Figure 6. The cross section Σ as a function of the reciprocal of the NaCl target temperature. k_i is about 7.7Å^{-1} and the deposition times range between 60 and 200 seconds per monolayer. Some estimates of errors in the measurements are given for several points.

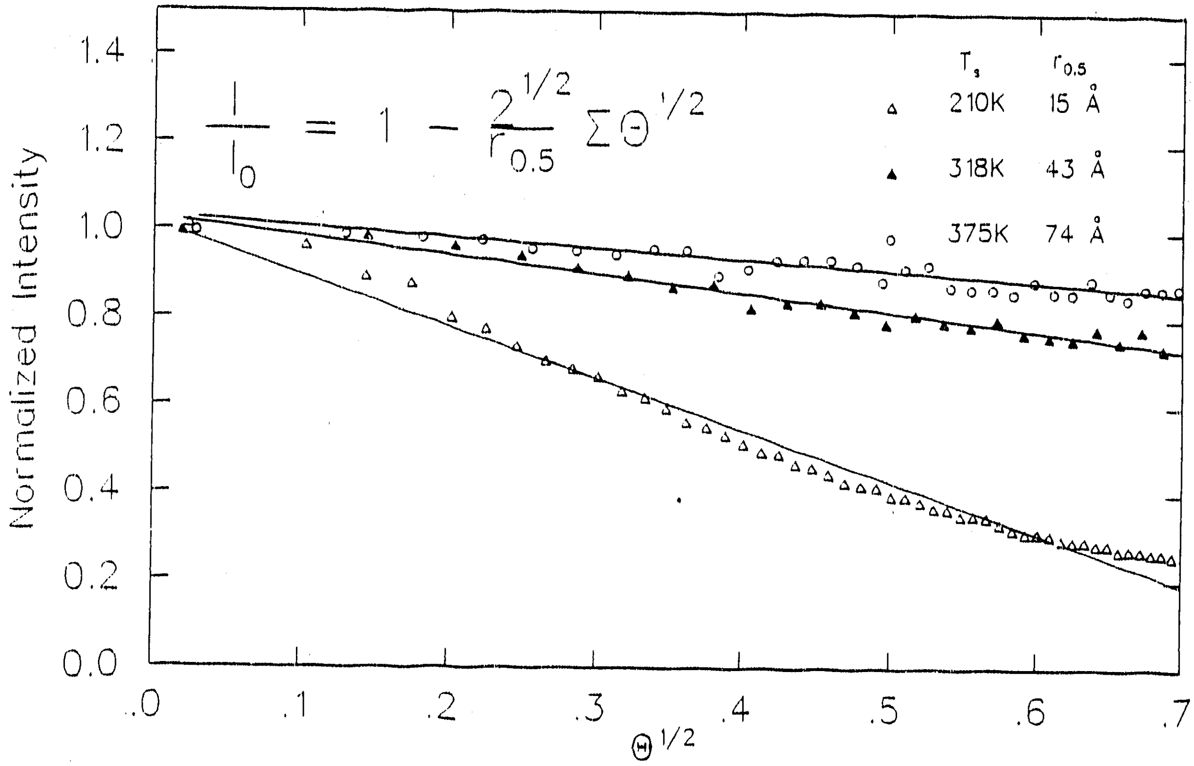


Figure 7. Plot of the specular beam intensity vs. $\Theta^{1/2}$ as in Equation (4) at 210K, 318K and 375K. In the experiments k_i = 7.7Å^{-1} and the value for Σ is taken to be 13Å (Ref. 14). The island radii, r_o, determined from the slopes are 15Å, 43Å and 74Å, respectively.

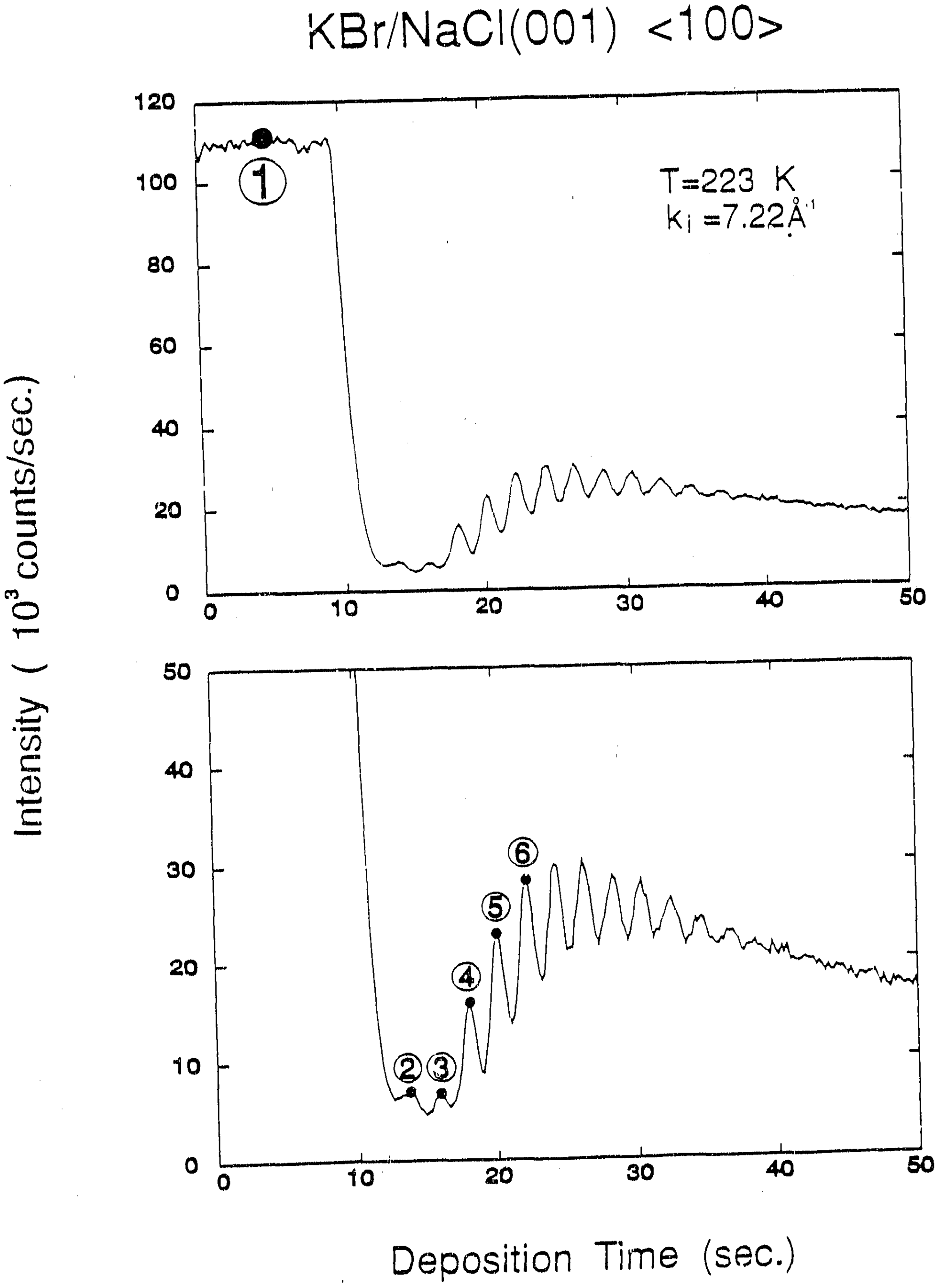


Figure 8. Deposition curve for KBr onto NaCl (001) <100>. The lower panel has the vertical scale expanded to display the oscillations better. The temperature of the surface is 223K and the initial He atom wavevector is 7.22Å^{-1}.

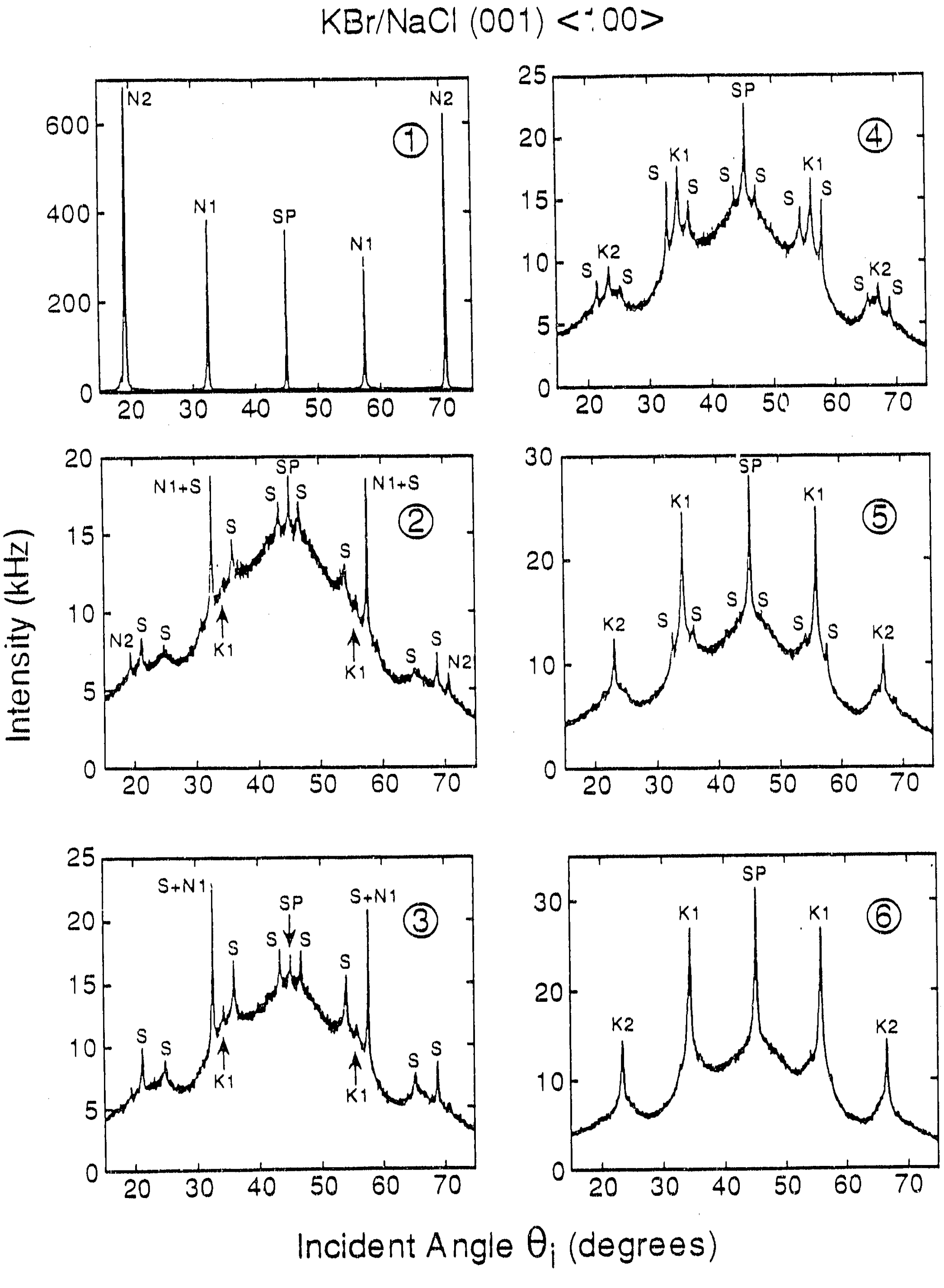


Figure 9. A series of angular distributions measured at the depositions indicated in the lower panel of Figure 8 in the <100> direction. In each case the surface temperature is 223K and the He atom wavevector is about 7.4Å^{-1}. The peaks in each panel are labeled according to the scheme: SP for the specular peak; N, K, and S for the Bragg diffraction peaks of NaCl, KBr and the superstructure, respectively. For NaCl and KBr, the additional numerical label 1 or 2 refers to the first or second order Bragg peaks, (11) and (22) at angles smaller than 45° and ($1\bar{1}$) and ($2\bar{2}$) at angles greater than 45°.

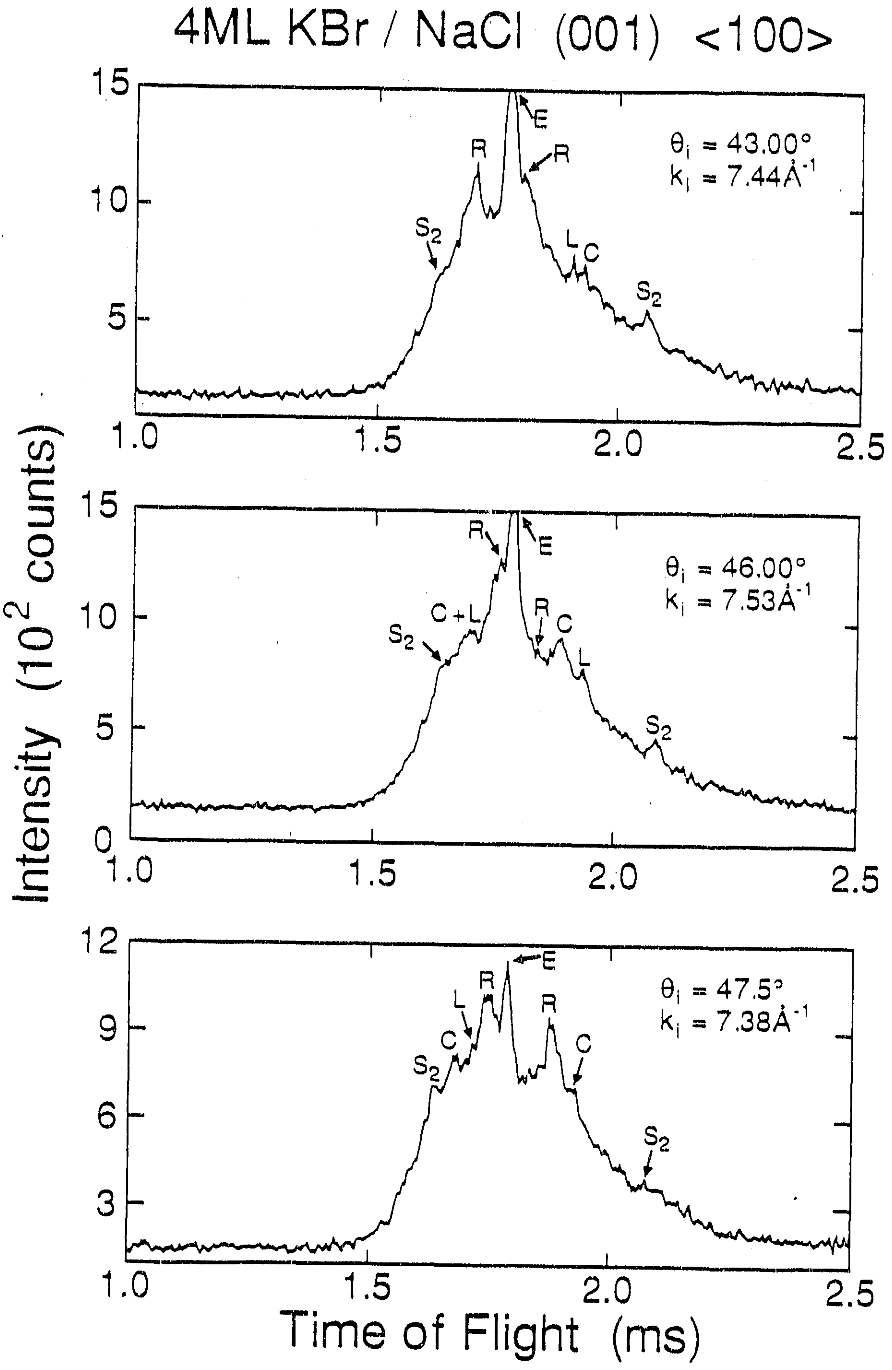


Figure 10. A series of representative time-of-flight spectra measured in the <100> direction. The surface temperature in each case is 223K; the angles and He atom wavevectors are given in each panel. The labels for the peaks are discussed in the text. Peaks lying to the right of the diffuse elastic peak correspond to surface phonon creation; those to the left correspond to annihilation. The data collection time for each spectrum is about 2 hours.

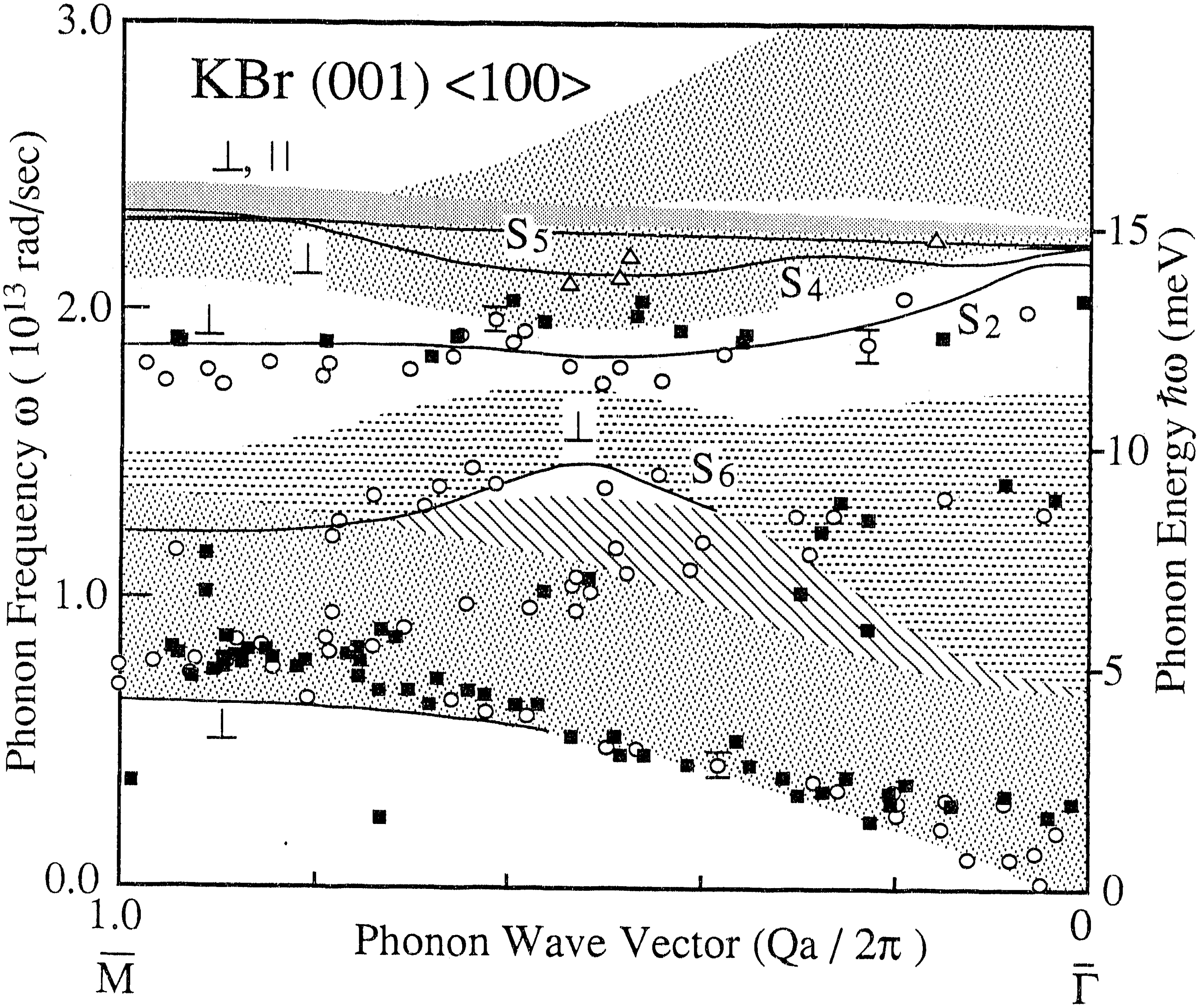


Figure 11. Surface phonon dispersion curves for KBr (001) in the < 100 > high symmetry direction. The open circles are points that were obtained previously using a cleaved KBr crystal surface; the open triangles were also obtained from the cleaved crystal but were of lower quality than the open circle data. The solid squares are points from the 4 monolayer surface of KBr on NaCl (001). The shaded regions and solid lines are the bulk bands projected onto the surface and the surface localized modes calculated by Benedek and Miglio.

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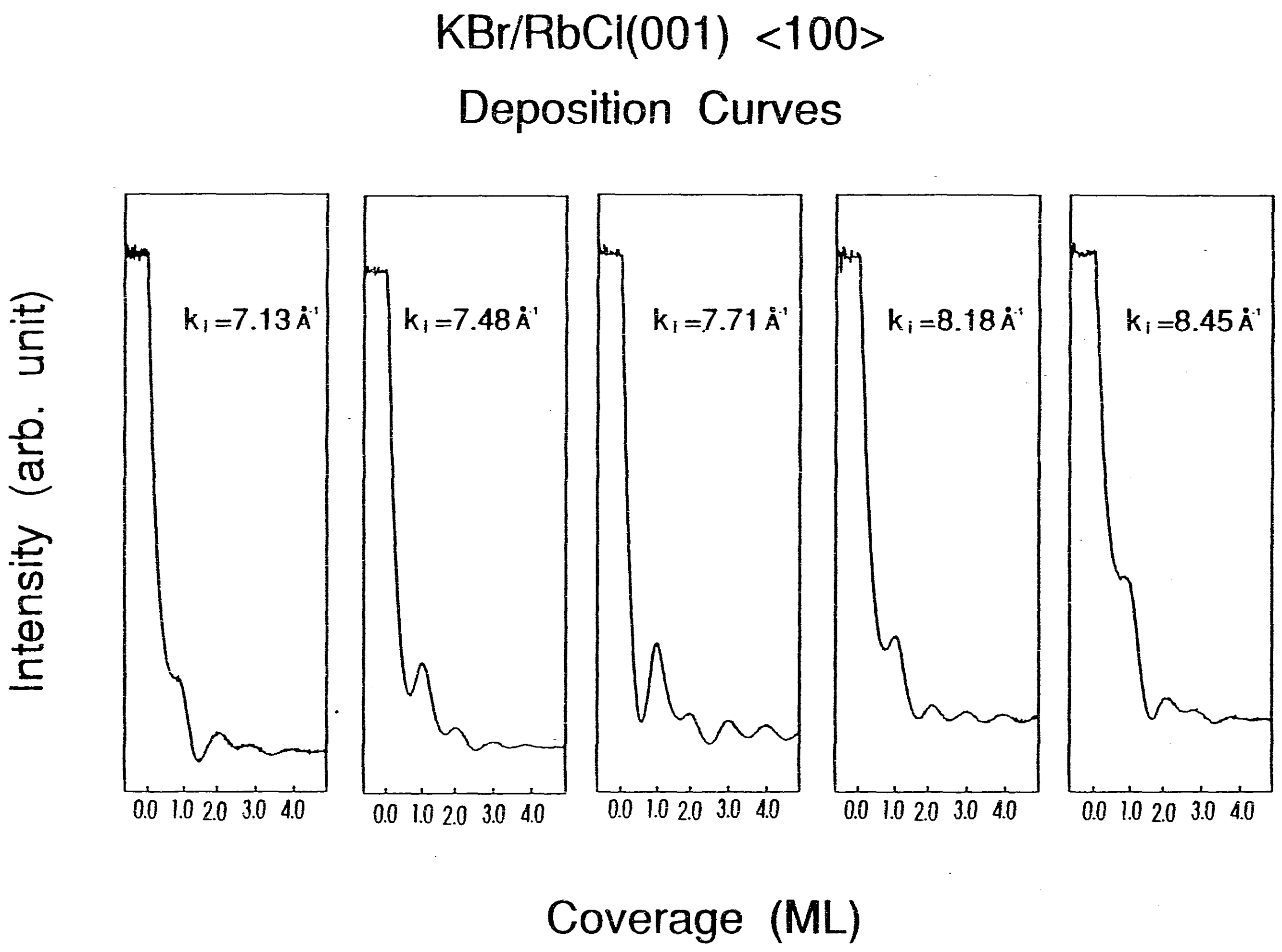

Figure 12. Plots of the specular intensity vs coverage θ in monolayers (ML) for different incident wavevectors k_i. The crystal temperatures are in the range of 190-210K for the measurements. The oscillatory pattern for the first monolayer has a different Δk_z dependence than for the subsequent ones. The center panel corresponds approximately to an out-of-phase scattering condition for the first monolayer and hence the deep first minimum, while the first and last panels are close to the out-of-phase condition for the second and subsequent monolayers. From the intensity oscillations of the first monolayer with Δk_z we calculate a KBr-RbCl step height of D_1=3.18Å. The second monolayer intensity oscillation gives for the KBr-KBr growth a value of D_2=3.46Å.

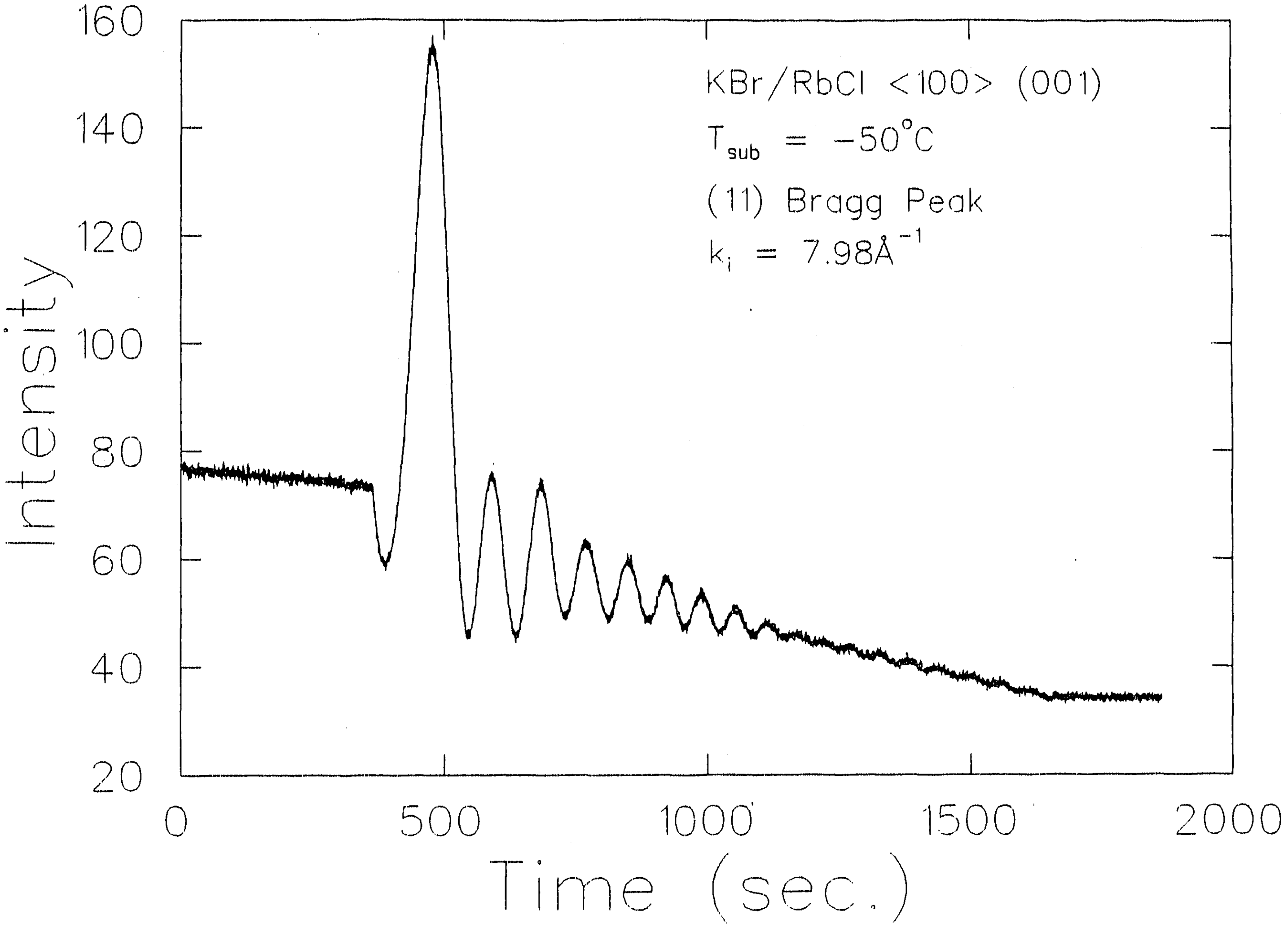

Figure 13. Plot of the intensity of the first order Bragg peak versus time (coverage) for KBr growing on RbCl(001) <100>. The initial large intensity is believed due to an enhancement of the corrugation in the first monolayer due to the mismatches in the ionic radii of the cations and of the anions even though the lattice dimensions are very nearly the same. This effect is very much reduced for subsequent monolayers, which then appear to grow very much as in homoepitaxy.

V. FACILITIES AND PERSONNEL

The support facilities for this project include the Machine and Electronic shops of both the Physics and Chemistry Departments. The employees of these shops are paid by the State of Florida and are normally available on a first-come-first-serve basis. In addition, the Center for Materials Research and Technology (MARTECH) which the State has set-up and funded at FSU, has technical and scientific personnel who are experienced in growing and handling crystals and thin films, and in operating an ESCA surface analysis station. More recently, a program of Molecular Beam Epitaxy has been developed here to make and study magnetic superlattices. In addition, there are facilities for X-ray, scanning electron microscope, ellipsometric and i.r. examination of crystal samples which are readily available to us on a limited basis. A MOCVD facility is now operating in the Chemistry Department. With the location of the new National High Field Magnetic Laboratory to be in Tallahassee, additional facilities and expertise is becoming locally available.

The Florida State University also has established the DOE-FSU sponsored Supercomputer Computations Research Institute (SCRI) which has a Cray Y-MP and a highly parallel Connection Machine. This facility is very well suited for collaborative ventures between our group and visiting theorists. (See below.)

The current scientific personnel consist of the co-principal investigators, JGS (Physics Department) and SAS (Chemistry Department), and five dedicated graduate students, J. Baker and J. Hernandez, G. G. Bishop, E. S. Gillman and J. Duan (all in Physics). Our first two students G. Chern (Physics) and W. P. Brug (Chemistry) have completed their doctoral dissertations. Chern is currently being paid by a DARPA grant to this institution as a post-doctoral associate doing MBE experiments and also working on transport properties of materials. Brug is working with the EPA in Montgomery, Alabama. Duan has just finished the growth work described in this proposal for his Ph.D. dissertation. Bishop is a fourth year student and has worked on some of the data analysis, particularly with temperature-dependent, multiphonon experiments. He also will be doing the surface lattice dynamics and density of states calculations when we develop our temperature dependent models further. He has begun experimental work recently on further alkali halide growth studies. Gillman is a third year student and has been working on characterizing a stainless steel alloy crystal and an α-Fe(110) crystal. He has developed the new alignment and polishing methods mentioned earlier, which improve greatly the ability to work with vicinal surfaces. He has also built a new high temperature target holder for the metal crystals. Hernandez and Baker are second year students who spent most of the past summer building a source to produce buckminsterfullerene (C_{60}) and probably will begin the alkyl thiol preparation and characterization experiments in the Spring. An undergraduate student (Physics), Ben Barnes is now assisting with the C_{60} work. Currently, three students are supported by the current grant from the U.S. Department of Energy, Duan is being supported by MARTECH and Baker has a Florida fellowship which will end next year.

Dr. Betty Derrick of Valdosta State College, Georgia, has spent a summer in our laboratory (1987) under a grant from the DuPont Foundation to the Chemistry Department and for various other shorter periods when her teaching schedule has permitted it. She applied for and has been awarded an NSF "Visiting Professorship for Women" grant to spend the next year (starting June 1992) working full time on the alkyl thiol/gold system.

In the past, we have had a number of undergraduates in our laboratory for periods of a semester through the Chemistry Department Honors Program and for summers through the Undergraduate Research Participation grants from the NSF to the Physics Department.

VI. COLLABORATIONS

We have had a long-standing collaboration with the MPI group in Göttingen, directed by Professor J. Peter Toennies. One of us (JGS) had a NATO grant to carry out experiments at the MPI, which has resulted in about a dozen published papers. More recently, we (JGS & SAS) have been awarded another NATO grant for further cooperative efforts between our group, the Göttingen group and the theory group of Benedek and Miglio in Milan, Italy. During next year Professor Toennies will spend a period of time with us.

We have a collaboration with Dr. Lynn Boatner at the Oak Ridge National Laboratory. Financial support for this work, mostly for travel, was awarded by the Southeastern University Research Association (SURA). It enabled our student, Mr. Gillman, to learn more about metal crystals and their preparation at Oak Ridge.

We have also collaborated at FSU with two Italian theorists, Professors Giorgio Benedek and Leo Miglio of the University of Milan and also their former student, Dr. Luciano Colombo who is now also on the faculty in Milan. Thanks to time on the SCRI computers provided by the Material Science Division of DOE, and financial assistance provided by SCRI and MARTECH at FSU, they were able to spend two months here (1988) to calculate the lattice dynamics of some metal and semiconductor superlattice materials. In the summer of 1989, Dr. Colombo was able to return to FSU for three weeks to carry out additional calculations. We expect to have Dr G. Onida from that group again this next year in association with the NATO grant. This work and its future extensions will be of great value to us in building a suitable theoretical understanding of our epitaxial growth experiments.

To interpret and understand the results of HAS experiments requires close collaboration with theorists. As an example of this, we note the collaborative effort in the interpretation and understanding of the crossing modes in the alkali halides with Professor Giorgio Benedek and in the temperature-dependent, multiphonon scattering model with Professor J. R. Manson of Clemson University. We intend to remain associated, in addition, with Professor Celli at the University of Virginia, a pioneer in surface-scattering theory, who is currently helping us understand the buckling behavior of our KBr/NaCl(001) layers and Professor Gerber of the Hebrew University, Israel, who is interested in surface scattering phenomena. Further, Professor F. W. de Wette from the University of Texas-Austin, a pioneering surface lattice theorist and a strong proponent of surface studies on the alkali halides and other ionic insulators, is very much interested in our new results for RbI.

VII. ACTIVITIES OF THE INVESTIGATORS

The principal effort of both of us has been to extend the techniques of He atom scattering, which have proven so successful as a probe of the dynamical behavior of surfaces, to the investigation of crystal growth. We have built on our experience with the alkali halides to study homo- and heteroepitaxy in this family of materials and plan to continue this effort and to expand into other insulators families. The construction of the instrument, except for the minor changes noted above, is complete and we are very pleased with the results so far.

In addition, we and our students have been active in the surface science community this year, having attended and given papers at various meetings. These include 1) the 1991 APS March meeting held in Cincinnati, 2) the Gordon Research Conference "Dynamics of Gas-Surface Interactions" in New Hampshire, 3) International Conference on Molecular Beams-Surface Interactions in Jülich, Germany, 4) the Fourth Eastern Regional Conference on Crystal Growth and 5) the 1991 national meeting of the American Vacuum Society held in Seattle. Further, we have presented papers at several regional meetings. We have also submitted abstracts for papers to be presented at meetings early next year and have been invited to present a paper on our recent work at the XIVth International Symposium on Molecular Beams to be held in Asilomar, California, in June, 1992.

Finally, both investigators have worked at the MPI in Göttingen this year in connection with the NATO grant, which resulted in a paper on C_{60}.

A. Papers Published or In-Press Since May, 1990.

1. "Surface Lattice Dynamics of the RbI(001) Crystal Surface via Inelastic He Atom Scattering", S. A. Safron, W. P. Brug, G. G. Bishop, J. Duan, G. Chern, and J. G. Skofronick, J. Elect. Spectros. Related Phenom. 54/55, 343-352 (1990).

2. "Surface and Bulk Phonon-Assisted Resonances with Surface Bound States in the Inelastic Scattering of He Atoms on NaCl(001)", S. A. Safron, W. P. Brug, G. G. Bishop, G. Chern, M. E. Derrick, J. Duan, M. E. Deweese and J. G. Skofronick, J. Vac. Sci. Technol. A9, 1657-1662 (1991).

3. "Surface Properties of CsF by He Atom Scattering", J. G. Skofronick, J. Duan, G. Chern, W. P. Brug, G. G. Bishop and S. A. Safron, Surf. Sci. 251/252, 782 (1991).

4. "Homoepitaxial Growth Investigated by High-Resolution He Atom Scattering: NaCl onto NaCl(001)", J. Duan, G. G. Bishop, E. S. Gillman, G. Chern, S. A. Safron and J. G. Skofronick, Accepted by J. Vac. Sci and Technol., Oct., 1991.

5. "Epitaxial Growth of KBr onto NaCl(001) by High Resolution He Atom Scattering", J. Duan, G. G. Bishop, E. S. Gillman, G. Chern, S. A. Safron and J. G. Skofronick, Accepted by Surf. Sci., Dec., 1991.

6. "Surface Lattice Dynamics of NiO(001) by Inelastic He Atom Scattering", W. P. Brug, G. Chern, J. Duan, G. G. Bishop, S. A. Safron and J. G. Skofronick, Accepted by J. Vac. Sci. Technol., Dec., 1991.

7. "Study of the Epitaxial Growth of Single Crystal C_{60} on Mica by Helium Atom Scattering", D. Schmicker, S. Schmidt, J. G. Skofronick, J. P. Toennies and R. Vollmer, Phys. Rev. B44, 10995 (1991).

B. Papers and Posters Presented in 1991 and Already Submitted for Early 1992.

1. "A Time-Resolved Study of an Electromagnetic Pulsed Nozzle Beam", E. S. Gillman, J. G. Skofronick, G. G. Bishop, W. P. Brug, J. Duan and S. A. Safron, 20th Annual Symposium on Applied Vacuum Science and Technology, Florida Chapter of the American Vacuum Society, February, 1991.

2. "Homoepitaxial Growth of NaCl on NaCl (001) Single Crystal Surface by He Atom Scattering", J. Duan, J. G. Skofronick, E. S. Gillman, G. Chern, W. P. Brug, G. G. Bishop and S. A. Safron, 20th Annual Symposium on Applied Vacuum Science and Technology, Florida Chapter of the American Vacuum Society, February, 1991.

3. "He Atom Studies of the Homoepitaxial Growth of NaCl on NaCl (001) Crystal Surface", J. G. Skofronick, J. Duan, G. Chern, W. P. Brug, G. G. Bishop and S. A. Safron, American Physical Society, national meeting, Cincinnati, Ohio, March, 1991.

4. "A Pedestrian View of Bucky-Balls", J. G. Skofronick, Max-Planck-Institut für Strömungsforschung, Göttingen, Germany, June, 1991.

5. "Investigation of Homoepitaxial Growth of NaCl onto NaCl (001) by He Atom Scattering", S. A. Safron, J. Duan, G. G. Bishop, E. S. Gillman, G. Chern and J. G. Skofronick, Gordon Research Conference "Dynamics of Gas-Surface Interactions", New Hampshire, August, 1991.

6. "He Atom Scattering Study of Layer-by-Layer Growth of NaCl on NaCl(001), J. Duan, G. G. Bishop, E. S. Gillman, G. Chern, S. A. Safron and J. G. Skofronick, The Fourth Eastern Regional Conference on Crystal Growth, American Association for Crystal Growth, Atlantic City, October, 1991.

7. "Epitaxial Growth of KBr onto NaCl (001) by High-Resolution He Atom Scattering", S. A. Safron, J. Duan, G. G. Bishop, E. S. Gillman, G. Chern and J. G. Skofronick, International Conference on Molecule-Surface Interactions, Jülich, Germany, October, 1991.

8. "Homoepitaxial Growth Investigated by High-Resolution He Atom Scattering: NaCl onto NaCl (001)", J. Duan, G. G. Bishop, E. S. Gillman, G. Chern, S. A. Safron and J. G. Skofronick, American Vacuum Society, national meeting, Seattle, Washington, November, 1991.

9. "Surface Lattice Dynamics of NiO (001) by Inelastic He Atom Scattering", W. P. Brug, G. Chern, J. Duan, G. G. Bishop, S. A. Safron and J. G. Skofronick, American Vacuum Society, national meeting, Seattle, Washington, November, 1991.

10. "Lattice Dynamics Study of KBr Epi-Layer on NaCl (001) by He Atom Scattering, J. Duan, G. G. Bishop, G. Chern, E. S. Gillman, S. A. Safron and J. G. Skofronick, Southeastern Section of the American Physical Society, Durham, N.C., November, 1991.

11. "Multiphonon Scattering of He Atoms from the KBr(001) and CsF())1) Surfaces", G. G. Bishop, J. G. Skofronick, J. Duan, W. P. Brug, S. A. Safron and E. S. Gillman, Southeastern Section of the American Physical Society, Durham, N.C., November, 1991.

12. "Surface Mobility of NaCl Adsorbates on the NaCl (001) Surface by Helium Atom Scattering, E. S. Gillman, G. G. Bishop, G. Chern, J. Duan, S. A. Safron and J. G. Skofronick, Southeastern Section of the American Physical Society, Durham, N.C., November, 1991.

13. "Production, Separation and Characterization of C_{60} and C_{70}", J. Baker, B. Barnes, J. Hernandez, S. A. Safron and J. G. Skofronick, 21th Annual Symposium on Applied Vacuum Science and Technology, Florida Chapter of the American Vacuum Society, February, 1992.

14. "Heteroepitaxial Growth Study of KBr/RbCl(001) via He Atom Scattering", J. Duan, G. G. Bishop, E. S. Gillman, G. Chern, S. A. Safron and J. G. Skofronick, American Physical Society, national meeting, Indianapolis, Indiana, March, 1992.

15. "Homo- and Heteroepitaxial Growth on Ionic Insulators via Helium Atom Scattering", S. A. Safron, J. Duan, G. G. Bishop, E. S. Gillman, G. Chern and J. G. Skofronick, XIVth International Symposium on Molecular Beams, (invited), Asilomar, California, June, 1992.

C. Proposals and Grants Other than DOE.

1. "Structure and Dynamics of Chemisorption through He Atom Scattering on Crystalline Surfaces", National Science Foundation Visiting Professorship for Women, with M. E. Derrick, June, 1992- February, 1993, funded $96,000.

2. "Dynamics of Crystal Surfaces, Both Clean and with Adsorbates", NATO, with J. P. Toennies, 1990-1991, funded $5447. Currently applying for a renewal for 1992-1993.

3. "FSU/ORNL Cooperative Research Program in Surface Science Studies", SURA, Summer, 1991, funded $5,880.

4. "Surface Dynamics of Insulators and Crystal Growth via High Resolution He Atom Scattering", National Science Foundation, July 1, 1992-June 30, 1995, $271,078, pending.

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