Origin and structure of streaks in reflection high-energy electron diffraction experiments

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An expression for one-phonon inelastic scattering of a high-energy electron beam has been obtained which is different from those of earlier workers. Numerical results for Ag(111) surface are very compatible with the recent experimental results for total (elastic plus inelastic) intensity. The structure seen in the streaks originates mainly from the penetration of the electron beam in the bulk.

I. INTRODUCTION

In the last decade, reflection high-energy electron diffraction (RHEED) has been seen to be quite promising in determining the configurations of the surface atoms on clean surfaces,[1–3] monolayer adsorbed surfaces,[4–6] or stepped surfaces.[7,8] In these studies, one should have a detailed understanding of the intensity distributions of the scattered beams before one may ascertain a particular atomic arrangement. Some progress has been made in the theoretical understanding[9–11] of RHEED patterns.

In diffraction experiments both elastic and inelastic scatterings are observed, and there appear streaks in the scattered distribution. A major contribution to the inelastic part comes from electron-phonon scattering. It is, therefore, desirable to make a study of phonon-induced electron scattering. In this paper, we investigate the thermal diffuse scattering associated with the electron scattering and obtain an analytical expression for one-phonon scattering which is different from that of earlier workers.[9–11] We then present numerical results for one-phonon inelastic scattering for the Ag(111) surface on which experiments have recently been performed. The experimentally observed streaks arise from inelastic scattering involving phonons, and the structure within the streaks is a consequence of the penetration of the electron beam.

II. INELASTIC ONE-PHONON SCATTERING

The scattering cross section for an electron of mass m scattered from an initial state $|\vec{k},i\rangle$ to a final state $|\vec{k}-\vec{K},f\rangle$ with a phonon emission of frequency ω and wave vector $\vec{K}$ is given by

$$\frac{d^2\sigma}{d\Omega\,d\omega}=\hbar\left[\frac{E_i+\hbar\omega}{E_i}\right]^{1/2}\left[\frac{m}{2\pi\hbar^2}\right]^2 \times|\langle\vec{k}-\vec{K},f\,|\,V\,|\,\vec{k},i\rangle|^2 \times\delta(\hbar\omega+E_i-E_f)\;, \tag{1}$$

where $|\vec{k},i\rangle$ denotes the combined state of energy E_i of the target state $|i\rangle$ and an electron of wave vector $\vec{k}$; V is the interaction potential and the other symbols have their usual meanings. Integrating over a small energy spread accounting for the instrumental effects one obtains a quasielastic cross section (static approximation)

$$\left[\frac{d\sigma}{d\Omega}\right]_{\rm qe}=\frac{1}{2\pi}\left[\frac{m}{2\pi\hbar^2}\right]^2\sum_{m,n}V_m^\dagger(\vec{K})V_n(\vec{K})e^{-W_m(\vec{K})}e^{-W_n(\vec{K})}e^{-i\vec{K}\cdot(\vec{R}_m-\vec{R}_n)}\exp|\langle[\vec{K}\cdot\vec{u}_m(0)][\vec{K}\cdot\vec{u}_n(0)]\rangle|\;. \tag{2}$$

Assuming that all of the atomic planes (including the surface) parallel to the surface are identical and that all of the atoms lying within a plane are similar, one may divide the solid into sets of planes designated by the index ν and exploit the two-dimensional translational symmetry parallel to the surface by writing

$$\vec{R}_m=\vec{d}_m+\vec{d}_\nu\;,\quad \vec{R}_n=\vec{d}_n+\vec{d}_{\nu'}\;,\quad \vec{K}=\vec{K}_{\parallel}+\vec{K}_z\;, \tag{3}$$

where $\vec{d}_m$'s are the atomic position vectors within the planes and $\vec{d}_\nu$'s denote the vectors lying along the third (z) axis *not* lying in the plane. The set $(\vec{K}_{\parallel},\vec{K}_z)$ represents the corresponding momentum-transfer wave-vector components. One may take into account the attenuation of the incident beam of electrons by a factor γ_ν which is the ratio of the amplitudes of the electron wave incident on the layer ν and on the surface. Equation (2) can then be written as

$$\left[\frac{d\sigma}{d\Omega}\right]_{\rm qe}=\frac{1}{2\pi}\left|\frac{m}{2\pi\hbar^2}V(\vec{K})\right|^2 e^{-2W(\vec{K})}\sum_{\nu,\nu'}\gamma_\nu\gamma_{\nu'}e^{-i\vec{K}_z\cdot(\vec{d}_\nu-\vec{d}_{\nu'})}\sum_{m,n}e^{-i\vec{K}_{\parallel}\cdot(\vec{d}_m-\vec{d}_n)}\exp|\langle[\vec{K}\cdot\vec{u}_m(0)][\vec{K}\cdot\vec{u}_n(0)]\rangle|\;. \tag{4}$$

We now represent the decrease in the amplitude of the electron wave incident on a particular layer below the surface by an analytical behavior, i.e., the amplitude decreases in a geometrical progression with layer by a factor γ. The different terms of the Taylor expansion of the exponential containing the correlation function $\langle[\vec{K}\cdot\vec{u}_m(0)][\vec{K}\cdot\vec{u}_n(0)]\rangle$ in Eq. (4) gives the cross section for electron scattering as a series of terms involving zero, one, and more phonons. The elastic scattering represented by the zeroth-order term is

$$\left[\frac{d\sigma}{d\Omega}\right]^{\text{el}} = \left[\frac{2\pi}{a}\right]^2 N_{\parallel}\left|\frac{m}{2\pi\hbar^2}V(\vec{K})\right|^2 e^{-2W(\vec{K})}A(K_z)\sum_{\vec{G}_{\parallel}}\delta(\vec{K}_{\parallel}-\vec{G}_{\parallel}) , \tag{5}$$

where $A(K_z)$ is the familiar Airy function to be discussed later and $\vec{G}_{\parallel}$ denotes the vectors in the two-dimensional reciprocal lattice; $N_{\parallel}$ is the number of atoms in one plane having lattice vector a.

The inelastic scattering cross section, involving one phonon, can be written as

$$\left[\frac{d\sigma}{d\Omega}\right]^{\text{in}} = \frac{1}{2\pi}\left[\frac{m}{2\pi\hbar^2}V(\vec{K})e^{-W(\vec{K})}\right]^2 \sum_{v}\gamma^v e^{-i\vec{K}_z\cdot\vec{d}_v}\sum_{m,n}e^{-i\vec{K}_{\parallel}\cdot(\vec{d}_m-\vec{d}_n)}\langle(\vec{K}\cdot\vec{u}_m)(\vec{K}\cdot\vec{u}_n)\rangle . \tag{6}$$

A plane-wave expansion of the atomic displacements in the two-dimensional reciprocal lattice gives the one-phonon cross section as

$$\left[\frac{d\sigma}{d\Omega}\right]^{\text{in}} = \left|\frac{m}{2\pi\hbar^2}V(\vec{K})e^{-W(\vec{K})}\right|^2 \frac{(2\pi)^2}{2V_sM}\sum_{v}e^{-i\vec{K}_z\cdot\vec{d}_v}\gamma^v\sum_{\alpha,\beta}K_\alpha K_\beta\sum_{s}\left[\frac{n(\omega_s)}{\omega_s}\xi_\alpha^{*s}\xi_\beta^{s}\sum_{\vec{G}_{\parallel}}\delta(\vec{K}_{\parallel}+\vec{q}_{\parallel}-\vec{G}_{\parallel}) + \frac{n(\omega_s)+1}{\omega_s}\xi_\alpha^{*s}\xi_\beta^{s}\sum_{\vec{G}_{\parallel}}\delta(\vec{K}_{\parallel}-\vec{q}_{\parallel}-\vec{G}_{\parallel})\right] , \tag{7}$$

where M is the atomic mass, V_s is the area of the planar unit cell; the subscripts α,β denote Cartesian components; the summation denoted by s includes all the phonon modes corresponding to a phonon wave vector $\vec{q}_{\parallel}$. We write $s=\{\vec{q}_{\parallel},p\}$ where p now denotes all the modes belonging to different polarizations belonging to the wave vector $\vec{q}_{\parallel}$. $\xi_\alpha^s \equiv \xi_\alpha(\vec{q}_{\parallel},p)$ are the eigenvectors; $n(\omega_s)$ is the phonon distribution function given by

$$n(\omega_s) = |e^{\hbar\omega_s/k_BT}-1|^{-1} .$$

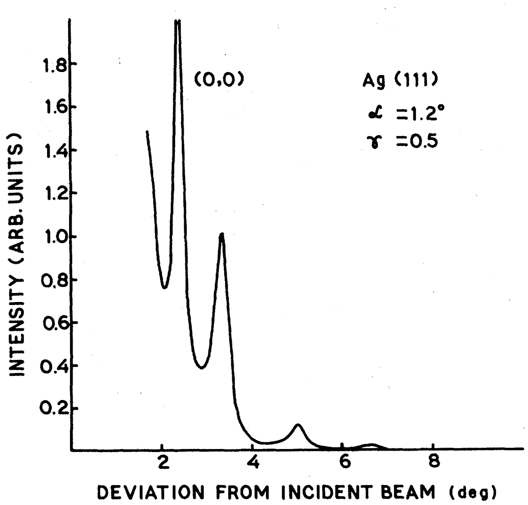

FIG. 1. Angular variation of the inelastic scattering cross section around the (00) rod of Ag(111) surface for an electron beam of 30 keV at an incident glancing angle (α) of 1.2° for $\gamma=0.5$.

As the evaluation of the expression (7) is quite complicated for a realistic crystal, we pass on to the high-temperature limit and assume a phonon Debye spectrum for obtaining an analytical expression. The validity of the approximation will be discussed later. In Debye approximation, one obtains

$$\left[\frac{d\sigma}{d\Omega}\right]^{\text{in}} = N_{\parallel}\frac{a}{\pi}\frac{k_BT}{Mc^2}K^2\left|\frac{m}{2\pi\hbar^2}V(\vec{K})\right|^2 e^{-2W(\vec{K})}A(K_z) \times \sum_{\vec{G}_{\parallel}}\tan^{-1}(q_{\max}^z/|\vec{G}_{\parallel}-\vec{K}_{\parallel}|)/|\vec{G}_{\parallel}-\vec{K}_{\parallel}| , \tag{8}$$

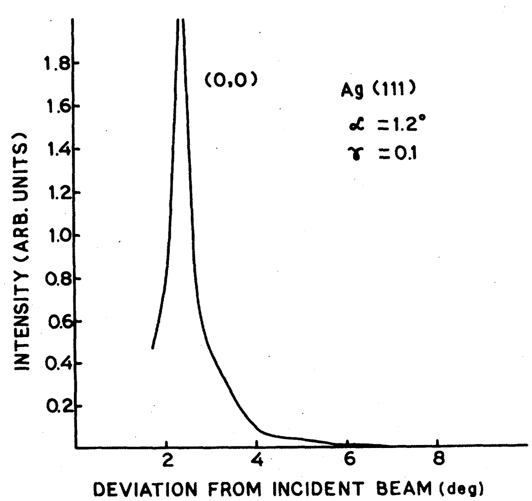

FIG. 2. Same as for Fig. 1, except that for $\gamma=0.1$.

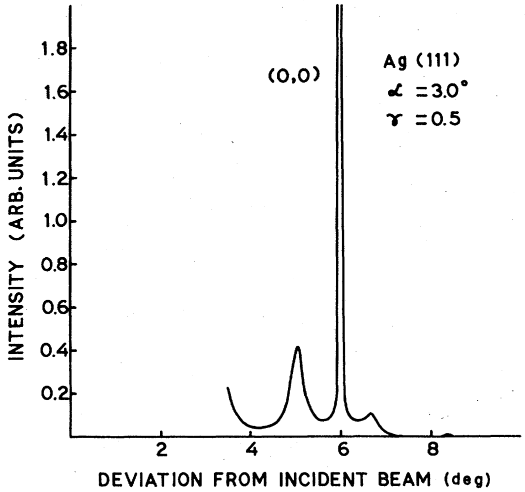

FIG. 3. Same as for Fig. 1, except that for an incident glancing angle of 3.0° and $\gamma=0.5$.

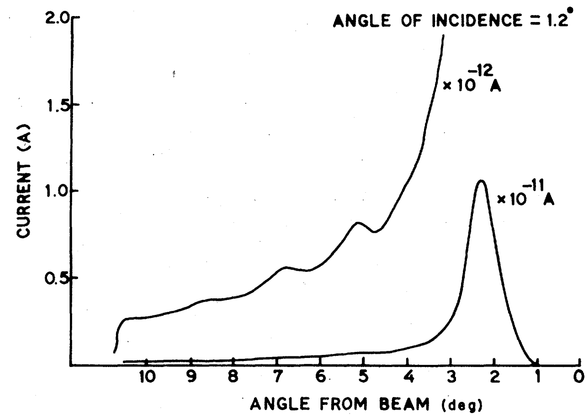

FIG. 5. Total (elastic and inelastic) intensity distribution for $\alpha=1.2°$ measured by Dobson.

with $q^z_{max} = | q^2_{max} - |\vec{G}_{\parallel} - \vec{K}_{\parallel}|^2 |^{1/2}$ and the Airy function $A(K_z)$ is given by

$$A(K_z) = | 1+\gamma^2 - 2\gamma \cos(K_z d) |^{-1} , \tag{9}$$

where d is the separation between the successive planes along the z axis; a is the lattice constant along the z axis; c is the averaged sound velocity in the solid.

An expression for inelastic scattering has been obtained earlier by Holloway and Beeby.[8] However, their expression contains an extra term inside the summation over $\vec{G}_{\parallel}$ in Eq. (8) in their effort to treat the infrared catastrophe. In fact, no such term appears as shown in the present calculation. Wallis and Maradudin[12] (WM) have earlier studied the one-phonon inelastic scattering of x-ray or low-energy electron diffraction from the surface *only* using a more realistic lattice model, i.e., a simple cubic lattice having central nearest and next-nearest interactions and have obtained an expression after making a numerical computation. The present scattering cross section is very similar to their expression except that the appearance of the Airy function $A(K_z)$ and a slightly different numerical factor [expand the summation over Cartesian components, regroup the terms in Eq. (42) of the WM paper, and replace $\tan^{-1}(\ )$ by $\pi/2$ in Eq. (8) of the present paper for a comparison].

The surface scattering determined mainly by the last factor $(1/|K_{\parallel}-G_{\parallel}|)$ will emerge as main peaks at $|K_{\parallel}-G_{\parallel}|=0$. We refer to them as the surface peaks in our future discussion.

The Airy function $A(K_z)$ appearing in Eq. (8) needs discussion. This factor takes into account the multibeam interference arising from the reflected electrons from the different layers of the solid. It may be rewritten as

$$A(K_z) = \left| (1-\gamma)^2 + 4\gamma \sin^2\left[\frac{K_z d}{2}\right] \right|^{-1} . \tag{10}$$

Thus, as is seen in the multibeam interferometry $A(K_z)$ will show structure in the scattering cross section. $A(K_z)$

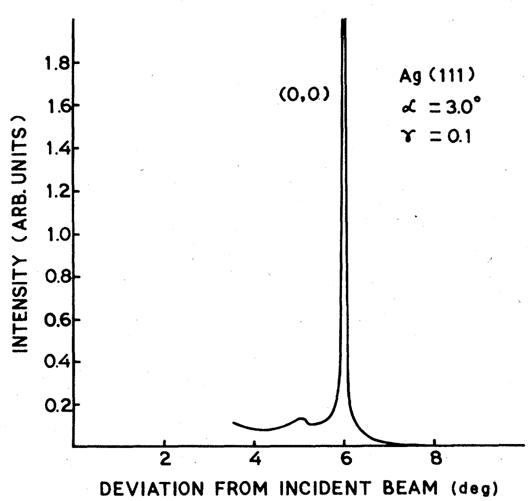

FIG. 4. Same as for Fig. 1, except that for an incident glancing angle of 3.0° and $\gamma=0.1$.

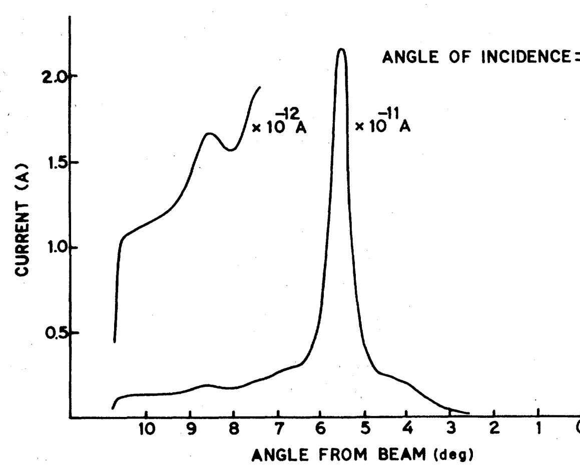

FIG. 6. Total (elastic and inelastic) intensity distribution for $\alpha=3.1°$ measured by Dobson.

will show maxima for values of K_z determined by

$$K_z d = 2n\pi \quad (n = 0,1,2,\ldots) . \tag{11}$$

The condition may be written as $d(\sin\theta+\sin\alpha)=n\lambda$, the familiar Bragg's reflection equation arising from the periodicity of the bulk solid in a direction perpendicular to the surface. Thus, $A(K_z)$ exhibits the bulk behavior. In a RHEED experiment the momentum transfer lying outside the surface, K_z, is quite large resulting in a number of maxima in the neighborhood of the surface peak. The widths of these peaks will depend on the attenuation factor γ. Greater is the penetration (high γ), sharper will be the peaks, and vice versa. In the limit of perfect penetration ($\gamma=1$), one obtains peaks similar to the Bragg peaks of the bulk crystal. In the other extreme situation, i.e., for a perfect nonpenetrating electron beam ($\gamma=0$), the scattering occurs at the surface only and one would see a smeared out variation of cross section away from the surface peak. One would not observe any structure in the inelastic cross section. In general, the intensity variation appears as streaks in the various RHEED experiments. One may easily extract information about the attenuation of electron beam by studying the structure in the inelastic scattering.

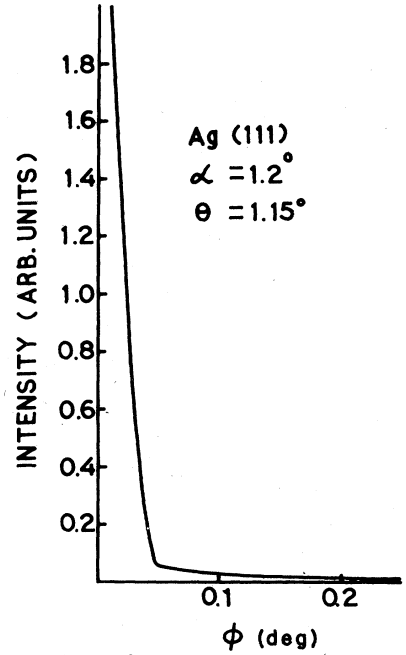

FIG. 7. Variation of inelastic scattering cross section for $\alpha=1.2°$ and $\theta=1.5°$ in a direction normal to the plane of incidence measured by angle ϕ.

III. CALCULATION AND RESULTS

The calculated variation of the inelastic scattering cross section around the surface peak arising from the (00) rod of the Ag(111) surface at $T=300$ K is shown in Figs. 1–4. The accelerating voltage of electron beam is chosen as 30 keV and the results are given for two values of $\gamma=0.5$ and 0.1. The values of the Fourier-transformed interaction potential $V(\vec{k})$ have been taken over from Doyle and Turner.[13] Figures 1 and 2 exhibit cross section for an electron beam incident at an angle $\alpha=1.2°$ and Figs. 3 and 4 for $\alpha=3.0°$. Two strong peaks appear on one side of the main peak corresponding to the (00) rod for $\alpha=1.2°$. However, for $\alpha=3.0°$ they appear on both sides of the main peak. The intensities of these extra peaks decrease as one moves away from the surface peak because of the Debye-Waller factor.

The above observed structures are seen to be in very good agreement with the recent measurements of Dobson[14] for the total elastic and inelastic scatterings (Figs. 5 and 6). The variation of the width of the streak normal to the incident plane is shown in Fig. 7. One notes comparatively a smaller spread along the normal direction.

In the above calculation we have assumed a Debye phonon spectrum which may not be appealing at the outset. Thus, we have determined the relative contributions of the phonons coming from the different parts of the Brillouin zone of a crystal. We observe that the main contribution to inelastic cross section comes from the immediate neighborhood of the center of the Brillouin zone. The contributions of the phonons having wave vectors away from the center is quite small. In fact, the cross sections remain practically unchanged even if one reduces the value of the maximum allowed phonon wave vector $q_{\max}$ by a factor of 4. One may, thus, safely assume a Debye spectrum for phonons in a RHEED theory when discussing a complicated situation.

In conclusion we find that the structures observed in the streaks in RHEED experiments arise mainly from the penetration of the electron beam and one may extract information about attenuation of electron beam from the analysis of these streaks.

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