Azimuthal dependence of the vibrational excitation in Si(111)-(2×1)

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The polarization of the surface vibrational excitation at 56 meV in Si(111)-(2×1) has been studied through high-resolution electron-energy-loss spectroscopy. By comparing the intensity of the loss peak for the incident plane normal and parallel to the chain direction of the surface reconstruction, we studied the surface anisotropy of the excitation. The experiments were also carried out at very low primary beam energies in order to avoid the masking effects of the kinematics. Comparing the results with the calculations performed with a model loss function, we obtained that both isotropic and anisotropic contributions are present in this loss feature.

Due to the 2×1 reconstruction mechanism the physical properties of the (111) surface of Si show a lower symmetry with respect to the ideal 1×1 surface. As a direct consequence of this, both the vibrational and electronic properties of the Si(111) surface are expected to show a strong dependence on the direction of the wave vector of the excitation in the surface plane (surface anisotropy).

A dramatic dependence of the surface electronic properties on the polarization of the exciting probe has been shown by surface optical spectroscopy with polarized light[1,2] and through high-resolution electron-energy-loss spectroscopy (HREELS).[3–5] Selloni and Del Sole[6] were able to describe, with a theoretical model, the main feature of the experimental findings in terms of the surface band structure.

A similar behavior has been proposed by Alerhand *et al.*[7–9] for the energy-loss peak due to the surface phonon excitation, observed at about 56 meV. However, on the experimental side, the evidence in favor of an azimuthal dependence of this surface mode is less clear and still under discussion. In fact, Di Nardo *et al.*[4] presented experimental evidence of the azimuthal dependence of the surface phonon by comparing their HREELS data as a function of the azimuthal angle with a phenomenological model[10] of the scattering due to an anisotropic surface phonon, in agreement with Alerhand *et al.*[7–9] In any case, they conclude that geometrical scattering parameters can mask the strong azimuthal anisotropy of the cross section when a parallel polarized phonon is excited.

We present here HREELS data in the region of the surface phonon excitation obtained in experimental conditions different from the conditions of Ref. 4. We show, on the contrary, an azimuthal dependence which cannot be explained by assuming only a single phonon mode (or branch) with eigenvector parallel to the chain direction of the reconstructed surface.

After a preliminary analysis of the azimuthal dependence of the cross section as a function of the scattering parameters, the scattering conditions have been chosen in order to emphasize the surface phonon anisotropy predicted by Alerhand *et al.*[7–9] The choice of scattering parameters has proved to be crucial in the possibility of making such anisotropy evident with HREELS. This critical behavior is a consequence of the scattering mechanism for electrons at the sample surface where, different from optical spectroscopy,[1] the incident probe can exchange a wave vector with a nonnegligible component in the direction normal to the plane of incidence and, even in nearly specular geometry, a surface mode with a wave vector with a nonzero component in the same direction can be excited.

Our results are close to the findings of Di Nardo *et al.*[4] when we use the same scattering condition, but they enable us to demonstrate, with different experimental parameters, the azimuthal dependence of the cross section and the contribution to the polarization of the phonon loss due to an isotropic component. Moreover, very recent calculations by Miglio *et al.*[11] improved the theory and also showed the presence of a component polarized normal to the surface at about 56 meV.

The paper is organized as follows. After describing the experimental setup, the scattering of electrons from an oriented surface dipole is discussed in the frame of a dipole interaction. The scattering conditions to put into evidence a possible surface anisotropy are then derived. Finally, the experimental results are presented and discussed.

High-resolution electron-energy-loss measurements

were performed by means of a Leybold-Heraeus ELS 22 spectrometer. The primary beam energy ranged between 0.85 and 19.7 eV, the energy resolution was 15 meV, and the incidence angle was 70°. The azimuthal dependence of the scattering efficiency was studied by rotating *in situ* the sample around the surface normal. Si samples were cleaved in a pressure less than 7×10^{-11} mbar (6.7×10^{-9} Pa) with a double-wedge–double-notch technique, and the presence of a single-domain (2×1) reconstruction on the whole surface as well as the chain direction were detected by low-energy electron diffraction (LEED). To avoid contamination, LEED measurements were performed after the electron-energy-loss spectroscopy measurements. Moreover, only those cleaves were chosen which showed (among a good reconstruction on the whole surface) a phonon and an electronic transition reliable in energy.[12]

The possibility of observing the anisotropy of a surface mode in HREELS in the dipole scattering regime is related to the scattering parameters. In particular, the scattering geometry and kinematics must be chosen in order to minimize the momentum transfer normal to the plane of incidence.

In fact, by simple kinematical considerations, it is shown that the ratio between the parallel (q_x) and the normal (q_y) component of the transferred two-dimensional wave vector $\mathbf{q}_\parallel=(q_x,q_y)$ is given by

$$\frac{q_x}{q_y}=A\frac{(E_p)^{1/2}}{(E_p-\hbar\omega)^{1/2}}+B\ ,$$

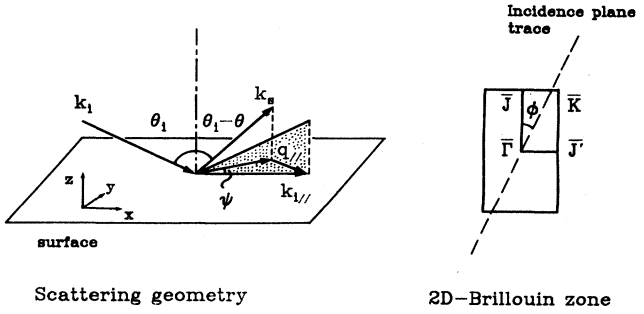
where

$$A=\frac{\sin\theta_i}{\sin(\theta_i-\theta)}\ ,$$

$$B=\frac{1}{\sin\psi}-\frac{\sin\theta_i}{\sin(\theta_i-\theta)}\ ,$$

$$q_x=(k_i-k_s)\sin\theta_i\sin\psi+k_s\sin(\theta_i-\theta)\ ,$$

$$q_y=k_s\sin(\theta_i-\theta)\sin\psi$$



Scattering geometry 2D–Brillouin zone

FIG. 1. Sketch of the principle of the experiment. The shaded plane represents the incidence plane. θ_i is the angle of incidence, θ and ψ are, respectively, the in-plane and off-plane angular deviations from the specular direction. ϕ is the azimuthal angle defined as the angle between incidence plane trace on the (111) surface and the $\bar{\Gamma}$-$\bar{J}$ direction of the surface Brillouin zone of the (2×1) reconstructed surface.

(see Fig. 1 for the meaning of the symbols). This simple result shows that q_x is much higher than q_y only when E_p is not too much higher than $\hbar\omega$. In the dipole scattering regime, the scattering efficiency is given by[13]

$$S_E=\frac{1}{RI_p}\int_{\Delta\omega}d\omega'\int_{\Delta\Omega_L}d\Omega_L\frac{m^2e^2v_\perp^2}{\pi\hbar^2\cos\theta_i}\left[\frac{k_s}{k_i}\right]\times\frac{4v_\perp^2q_\parallel^2}{[q_\parallel^2v_\perp^2+(\omega'-\mathbf{v}_\parallel\cdot\mathbf{q}_\parallel)^2]^2}\times g(\mathbf{q}_\parallel,\omega')\ , \quad (1)$$

where $g(\mathbf{q}_\parallel,\omega')$ is the surface loss function arising from surface vibration modes. We assume that the kinematical prefactor is not dependent on the azimuthal angle (ϕ).

Following a semiclassical description given by Mills,[14] the surface loss function can be written for a semi-infinite medium in terms of the polarization vector $\mathbf{P}=(\mathbf{P}_\parallel,P_z)$ arising from surface vibrations. If we assume $\mathbf{P}$ to be constant in the region of $bfq_\parallel$ involved in the experiments (near $\mathbf{q}_\parallel=0$) and we choose only vibrational modes with *even* symmetry with respect to the glide symmetry plane of the surface, $\mathbf{P}_\parallel$ is directed along the chains of Si(111) reconstruction and P_z is perpendicular to the surface. One has

$$g(\mathbf{q}_\parallel,\omega)=\left[\frac{4\pi ne\epsilon}{1+\epsilon}\right]^2\left[P_z-\frac{i}{\epsilon}\hat{\mathbf{q}}_\parallel\cdot\mathbf{P}_\parallel\right]^2\ , \quad (2)$$

where the azimuthal dependence can be due only to the second term.

The integration in Eq. (1) over the finite angles of acceptance of the spectrometer $\Delta\Omega_L$ introduces an average on $\mathbf{q}_\parallel$ vectors which can mask the effect of anisotropy. To obtain a quantitative evaluation of the relative scatter-

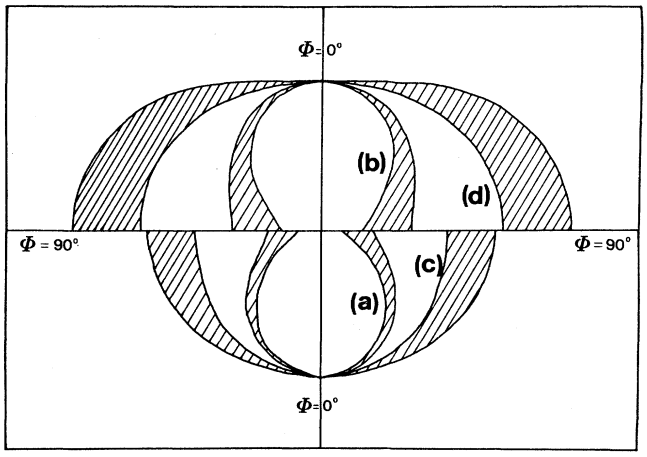


FIG. 2. Dependence of the scattering efficiency on the azimuthal angle, at $E_p=$(a) 0.85, (b) 1.53, (c) 7.20, and (d) 19.70 eV. The dashed area for each polar plot is the calculated uncertainty due to the varied Gaussian weight on the entrance slit of the analyzer and to the work-function difference between the electron-gun cathode and the sample.

ing efficiency for every azimuthal direction, we have integrated over the finite slit of our spectrometer ($\Delta\theta_s = 0.2°$, $\Delta\psi = 3.17°$) Eq. (1) weighted with a Gaussian of FWHM equal to the value of the angular aperture. We have also considered the most unfavorable value for the experimental parameters in order to obtain the limiting conditions to verify the anisotropy.

In particular, we have varied the Gaussian weight to simulate the transmission of the analyzer. We also considered the possible error (200 meV) on the estimation of the primary beam energy, taking into account the work-function difference between the electron gun cathode and the sample. All these parameters were chosen also in the analysis of the surface electronic transition[3,5] where we demonstrated the complete anisotropy of the excitation.

The results of the analysis, shown in Fig. 2, describe the dependence of the scattering efficiency on the azimuthal angle. As can be inferred from the figure, it is possible to test the anisotropy of the phonon excitation by choosing a very low primary beam energy, because the contribution of modes with $\mathbf{q}_{\parallel}$ component normal to the incidence plane is minimized. One can observe that when the primary beam energy increases, the anisotropy is masked and it is not possible to discriminate the polarization of the excitations. At very high impact energies the entrance slit of the electron analyzer can accept a large $\mathbf{q}_{\parallel}$ contribution normal to the plane of incidence, leading to an inversion of the intensity ratio if the excitation is polarized in the direction perpendicular to the plane of incidence.

In Figs. 3(a)–3(d) we show the HREEL spectra of the vibrational loss at about 56 meV of the Si(111)-(2×1) reconstructed surface collected at various primary beam energies E_p ranging from 0.85 to 19.7 eV. In the upper scale of each panel we report the exchanged wave vector $\mathbf{q}_{\parallel}$ corresponding to the conservation laws. To obtain quantitative information we extracted the loss functions from the experimental scattering efficiencies by dividing the spectra by the integrated kinematical prefactor. Only a mild anisotropy is present when the primary beam energies are low (a set of data taken at $E_p = 1.10$ eV, not presented in the figure for simplicity, is completely consistent with the data shown). If the energy of the incoming electrons increases, the apparent anisotropy is further reduced by the effect of a wider range of accepted q_y by the analyzer slit.

All the experimental parameters (low primary beam energy and grazing incidence angle) were chosen in such a way as to make evident the anisotropy of the phonon loss predicted by Alerhand *et al.*[7–9] To verify this behavior we report in Figs. 3(e)–3(h) the comparison between the azimuthal dependence of the loss functions calculated by the model previously described and the relative intensities of the phonon loss evaluated from the experimental loss functions at $\phi = 0°$ and 90° (the experimental intensity has been normalized to the theory at $\phi = 0°$).

At low primary beam energies there is not the expected strong reduction of the intensity, and when the ratio of the loss to the impact energy is decreasing we do not observe a strong inversion of the intensity. The wide range of scattering parameters is crucial to a better understand-

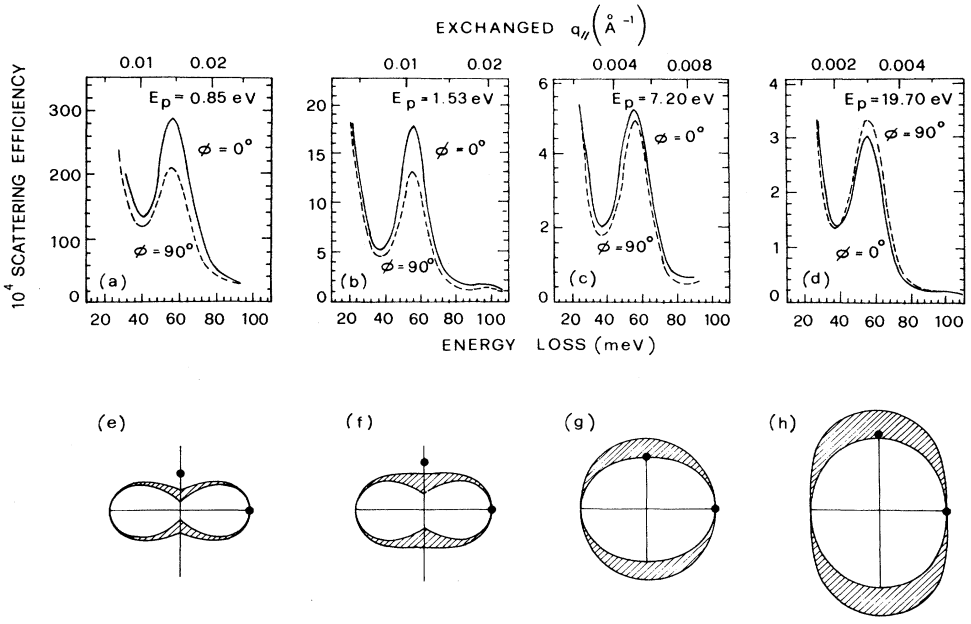


FIG. 3. Scattering efficiency is $I(\hbar\omega)/(RI_p)$, where $I(\hbar\omega)$ is the anelastic intensity, R the surface elastic reflection coefficient, and I_p the primary beam intensity, vs $\hbar\omega$ at different azimuthal angles at E_p = (a) 0.85, (b) 1.53, (c) 7.20, and (d) 19.70 eV for the vibrational excitation taken along $\phi = 0°$ (solid line) and $\phi = 90°$ (dashed line); in the upper scale of each panel the exchanged $q_{\parallel}$ values are indicated. At the bottom of the figure the comparison between the dependence of the loss functions on the azimuthal angle and the experimental ratio between the intensity of the vibrational loss at $\phi = 0°$ and 90° are indicated.

ing of the phonon polarization. Di Nardo *et al.*[4] performed the experiment only at a primary beam energy of about 7 eV with the analyzer slit of $\Delta\theta_s = 1°$ and $\Delta\psi = 1°$; they did not minimize the momentum transfer normal to the plane of incidence, and therefore they were not able to put into clear evidence the expected anisotropy.

In conclusion, by exploiting a wide range of primary beam energies, starting from very low values, we observe that the experimental results are not in agreement with the theory which predicts a straight parallel polarization along the chain direction, even accounting for limiting cases for the lateral acceptance of the analyzer. After the inclusion of all these conditions we can exclude that the geometrical scattering parameters are masking a complete azimuthal dependence of the cross section when a parallel polarized phonon is excited. This case is different from the one of the energy loss due to electronic excitation involving surface states at higher energies, where the full anisotropy is well established.[1–5]

Support for our experimental findings comes from a theoretical study of the Si(111)-(2×1) surface dynamics by Miglio *et al.*[11] They considered the crucial influence of the second layer to the dynamics of this extensively reconstructed surface. In particular they find, apart from the optical anisotropic vibration of the topmost chains, two "interface" vibrations at about 56 meV in $\bar{\Gamma}$ polarized perpendicular to the surface.

Therefore we are led to conclude that, at the energy loss of about 56 meV in Si(111)-(2×1) surface, an isotropical contribution to the cross section due to modes polarized along the surface normal coexists with the anisotropical contribution of a mode polarized along the surface *chains*.

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[1]P. Chiaradia, A. Cricenti, S. Selci, and G. Chiarotti, Phys. Rev. Lett. **52**, 1145 (1984).

[2]M. A. Olmstead and N. A. Amer, Phys. Rev. Lett. **52**, 1148 (1984).

[3]U. del Pennino, M. G. Betti, C. Mariani, C. M. Bertoni, S. Nannarone, I. Abbati, L. Braicovich, and A. Rizzi, Solid State Commun. **60**, 337 (1986).

[4]N. J. Di Nardo, W. A. Thompson, A. J. Schell Sorokin, and J. E. Demuth, Phys. Rev. B **34**, 3007 (1986).

[5]U. del Pennino, M. G. Betti, C. Mariani, C. M. Bertoni, S. Nannarone, I. Abbati, L. Braicovich, and A. Rizzi, Surf. Sci. **189/190**, 689 (1987).

[6]R. Del Sole and A. Selloni, Surf. Sci. **168**, 35 (1986).

[7]O. L. Alerhand, D. C. Allan, and E. J. Mele, Phys. Rev. Lett. **55**, 2700 (1985).

[8]O. L. Alerhand and E. J. Mele, Phys. Rev. Lett. **59**, 657 (1987).

[9]O.L. Alerhand and E. J. Mele, Phys. Rev. B **37**, 253 (1988).

[10]N. J. Di Nardo and E. J. Mele, Surf. Sci. **173**, L659 (1986).

[11]L. Miglio, P. Santini, P. Ruggerone, and G. Benedek (unpublished).

[12]J. A. Schaefer, J. Electron Spectosc. Relat. Phenom. **44**, 373 (1987).

[13]D. L. Mills, Surf. Sci. **48**, 59 (1975).

[14]D. L. Mills, Prog. Surf. Sci. **8**, 143 (1977).