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Lattice dynamics of the Ni(977) surface

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In a recent Letter, Mele and Pykhtin [Phys. Rev. Lett. **75**, 3878 (1995)] presented a calculation of the surface dynamics of the vicinal Ni(977) surface based on continuum elastic theory that they have used to explain recent inelastic He-atom scattering phonon data obtained by Niu *et al.* [Science **268**, 847 (1995)]. We have calculated the lattice dynamics of this surface in the framework of a microscopic force-constant model that gives the entire surface-phonon dispersion curves and provides detailed insight into the origin and character of the step-localized phonon modes. [S0163-1829(97)01904-8]

In a recent Letter Mele and Pykhtin[1] reported on a calculation of the surface dynamics of the Ni(977) surface based on continuum elastic theory, which they have used to explain recent inelastic He-atom scattering (HAS) phonon data.[2] Previously, the surface dynamics of stepped (metal) surfaces had been analyzed in several calculations including also the Green's function method.[3–5] In particular, the existence and nature of step-localized phonon modes has recently been demonstrated both experimentally and theoretically.[6,7] Mele and Pykhtin have used continuum elastic theory to explain some of the step phonon features observed for long wavelength close to the $\overline{\Gamma}$ point on Ni(977).

In this paper we wish to demonstrate that the phonon dispersion curves of stepped surfaces can also be calculated using a microscopic force-constant model, which provides the full information on the polarization of all modes, for all azimuths over the entire Brillouin zone. This is illustrated by applying the lattice dynamical theory described previously[7,8] to the Ni(977) surface. The calculations were performed for a slab of 100 layers (each layer containing all eight unit cell atoms) with the ideal atomic positions and a radial Ni force constant of $\beta=36.8$ N/m.[9] This simple force-constant model has been found to provide a reasonable first-order approximation for the bulk phonon dispersion curves of several metals and reproduces the values of the velocities of sound. Moreover, since the phonon frequencies are approximately proportional to $\sqrt{\beta/M}$, they can be adapted to other materials using the corresponding values of β/M. In previous work we found that we could fully explain the observed phonons on various stepped copper surfaces with a single force constant,[7] which has also been found to provide a good first-order approximation for the Ni surfaces.[10,11] Since our concern is to deduce the main lattice dynamical properties and classify the surface phonons of the Ni(977) surface we have at first neglected step relaxation effects. In a second step we have studied the effect of a step atom relexation on the phonon spectrum. The number of layers is found to be sufficient to decouple both slab surfaces since the modes close to the $\overline{\Gamma}$ point have the same frequencies to within 0.02 meV. We note further that the inclusion of a small tangential force constant does not change the character of the calculated surface modes.

Figure 1 summarizes the calculated dispersion curves for surface modes and resonances and their polarizations for the symmetry directions parallel ($\overline{\Gamma X}$) and perpendicular ($\overline{\Gamma Y}$) to the step edges. Since the length of the (977) surface unit cell along the $\overline{\Gamma Y}$ direction is approximately eight times larger than the corresponding (111) surface unit cell the surface phonon dispersion curve is given in a first approximation by the sevenfold folded surface phonon spectrum of the (111) surface. Along the $\overline{\Gamma Y}$ direction the Rayleigh mode and several backfolded branches were identified (solid lines in Fig. 1) with $\overline{Y}$-point ($\overline{\Gamma}$-point) energies of 3.2 meV (6.1 and 6.2 meV), 8.8, and 9.1 meV (11.3 and 11.4 meV), 13.3 and 13.5 meV (14.7 and 15.1 meV) and 15.6 meV, respectively. This folding is more pronounced than on the previous studied Cu(211) and Cu(511) surfaces[7] with much smaller terrace spacings. On the Ni(977) surface six of the eight terrace atoms (within the surface unit cell) have the same coordination as on the (111) surface (see inset in Fig. 1) and are therefore not so strongly affected by the steps. For this reason only small gaps of the order of 0.1 to 0.4 meV appear between the backfolded branches at the symmetry points, which result from the perturbation produced by the step edges. Additional backfolding with a small density of states

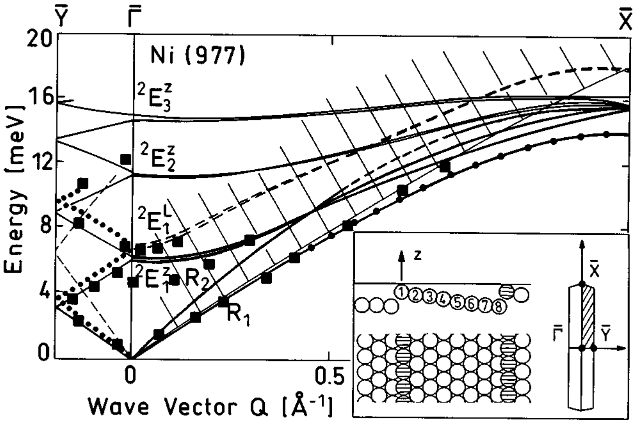

FIG. 1. Calculated surface phonon dispersion curves for both high symmetry directions of the Ni(977) surface. The dashed, dotted, and solid lines represent surface modes and resonances with longitudinal, horizontal, and *z* polarization, respectively. The squares are the experimental data obtained by Niu *et al.* (Ref. 2). The hatched area denotes the lower bulk band edge.

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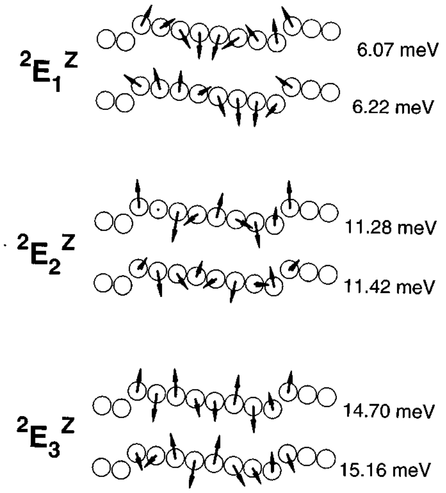

FIG. 2. Atomic displacement patterns for the different saggital polarized doublets at the $\overline{\Gamma}$ point. Corresponding to the simple, twofold, and threefold , folded Rayleigh mode branches along the $\overline{\Gamma Y}$ direction the $^2E_1^Z$, $^2E_2^Z$, and $^2E_3^Z$ doublets are characterized by 2, 4, and 6 nodes, respectively.

has been identified for the longitudinal resonance with a $\overline{Y}$-point zone boundary (zone center) energy of 6.2 meV (12.2 meV) and for the transversal resonance at 3.45 meV (6.7 and 6.8 meV) and 9.6 meV. The higher-order folded branches of these resonances could not be resolved presumably because they mix very strongly with the bulk bands.

Along the $\overline{\Gamma X}$ direction the lowest-energy surface mode is the saggital polarized Rayleigh mode (R_1) with an $\overline{X}$-point zone boundary energy of 14.1 meV. At the zone boundary this mode is found to be localized at the step edges with the largest displacement at the step atoms (see Fig. 3). A further (secondary) Rayleigh type resonance (R_2) with predominant terrace atom displacements was also observed. It lies mainly within the transversal bulk band and then appears at the $\overline{X}$ point where it has an energy of 15.65 meV. We note that at the $\overline{\Gamma}$ point the backfolded branches from the $\overline{\Gamma Y}$ direction of the Rayleigh mode and the horizontal resonance match to the z-polarized and longitudinal mode doublets, respectively, propagating along the step edges $\overline{\Gamma X}$. The sagittal polarized doublets ($^2E_1^Z$, $^2E_2^Z$, and $^2E_3^Z$) can be classified by the number of the nodes of their displacement patterns of the terrace atoms (see Fig. 2). The ability of the microscopic approach to predict the zone boundary displacement patterns is another advantage over the continuum theory. The dispersion curves of these step-induced saggitally polarized resonances pass through the bulk band and appear below the bulk band at the $\overline{X}$ point close to the R_2 Rayleigh mode at energies between 15.73 and 15.85 meV. At 0.85$\overline{\Gamma X}$ one of the $^2E_3^Z$ doublets acquires an increased transversal polarization and appears at the zone boundary at 16.43 meV as a further step localized mode (see Fig. 3).

A further step-induced longitudinal resonance doublet ($^2E_1^L$) lies below the longitudinal bulk band with $\overline{\Gamma}$-point energies of 6.7 and 6.8 meV. At the zone boundary no gap between the longitudinal and the transversal band and thus no longitudinal gap mode exists. However, two longitudinal resonances appear with $\overline{X}$-point energies of 30.5 and 31.6 meV.

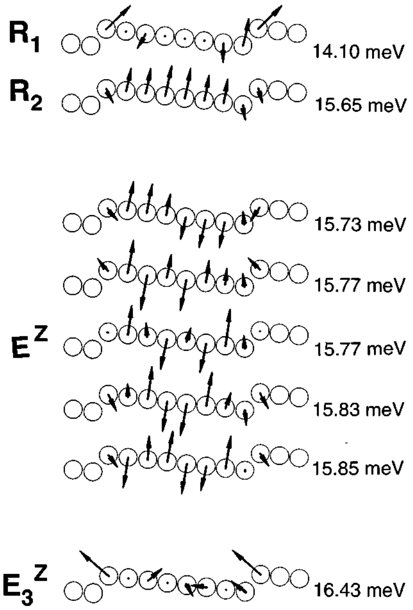

FIG. 3. Atomic displacement patterns for the saggital polarized modes at the $\overline{X}$ point. The modes with energies of 14.10 meV (R_1) and 16.43 meV (E_3^Z) are virtually step localized.

It is well known that the top-layer step atoms of vicinal metal surfaces relax inwards.[12] Usually, this relaxation is described in terms of a relative reduction of the interlayer spacing, Δ_{12}, which corresponds in our notation to a change in the z component of the spacing between the first and second atom within the surface unit cell (see Fig. 1). In order to study the relaxation effect on the surface phonons we have used typically a value of $\Delta_{12}=-20\%$ (which corresponds to a reduction of the distance to the underlying atoms by 2%) and increased the radial interlayer force constant according to the Badger's rule by 20%. This treatment was previously found to give a good description of the lattice relaxations on Ni(110) and Cu(110) surfaces.[13,14] Surprisingly, the calculation reveals that this change of the force constants does not significantly affect the zone boundary energies and gap widths within about 0.01 meV. Only at the $\overline{X}$ point did the step-edge-localized modes and the longitudinal resonance increase by 0.2, 0.4, and 1 meV, respectively. This insensitivity of most of the surface modes to local force field changes at the steps reflects the small contribution of the displacements of the step edge atoms — except for certain two edge localized modes (see Fig. 3).

For the $\overline{\Gamma X}$ direction Mele and Pykhtin reported a primary and secondary Rayleigh wave. Close to the $\overline{\Gamma X}$ point they found a step-induced doublet and a longitudinal bulk band resonance. Unfortunately, they have not shown the analysis of the surface modes propagating perpendicular to the steps ($\overline{\Gamma Y}$ direction) nor presented the higher-energy doublets that arise from the other backfolded branches. The present calculation provides all step-induced doublets at the $\overline{\Gamma}$ point and moreover clearly distinguishes between their different polarizations. The sagittal polarization of the $^2E_1^Z$ doublet is in agreement with the experimental assignment by Niu *et al.*[2] For the ${}^2E_1^l$ mode we find a longitudinal polarization in contradiction to their assignment to a mode of predominantly shear-horizontal character. Since both modes were observed with HAS without an accompanying umklapp processes[15] or tilting the sample[7] the excitation of a shear-horizontal polarized mode is forbidden by symmetry considerations.[16]

In order to compare the Rayleigh modes of a vicinal surface with those of the corresponding terraces the different geometries and the angles between the corresponding Brillouin zones have to be taken into account.[17] For comparison with the defolded branches along the $\overline{\Gamma Y}$ direction we have calculated using the same Ni force constant the Rayleigh mode of the Ni(111) surface along the corresponding $\overline{\Gamma M}$ direction. The complete agreement of both calculated Rayleigh mode dispersion curves indicates that we can exclude that the softening of the Ni(977) Rayleigh mode on the terraces of Ni(977) as observed by Sibener *et al.*[2] is merely caused by the reduced symmetry of this vicinal surface and must have another origin. Since the backfolded Rayleigh mode branch along the $\overline{\Gamma Y}$ direction must be matched to the step-induced doublets at the $\overline{\Gamma}$ point (see Fig. 1) the observed interlayer softening implies also a reduction of the ${}^2E_1^Z$ mode energy whereas the longitudinal mode (${}^2E_1^L$) is not affected. This is consistent with the observed softening, which implies that the zone center step mode energy for ${}^2E_1^Z$ must be reduced from 6 meV close to the measured value of 5 meV.[2] We note, however, that the experimental data (also shown in Fig. 1) reveal significantly different energies for the backfolded Rayleigh mode and the ${}^2E_1^Z$ doublet at the $\overline{\Gamma}$ point. Presently it is not clear whether this is due to an inadequacy of the model or experimental errors. Thus the surface should be reinvestigated with improved resolution. In particular, a knowledge of the X-point energies of the two step-localized modes would provide very important new information on the local force field at step edges and step atom relaxation.

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