

Observation of Pronounced Neon Diffraction from Low-Index Metal Surfaces

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Neon diffraction from dense metal surfaces is reported for the first time. Contrary to expectation, pronounced rainbow scattering at angles of $\sim 12^\circ$ for Ni(110) and $\sim 40^\circ$ for Pd(110) shows that the corrugation amplitudes are much larger for Ne than for He. This opens important extensions for atomic-beam studies of surface structure and composition and, as Ne is also more sensitive to dilute adsorbates, of surface defects and migration.

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The potential of He scattering for surface research has been explored successfully in the last few years:

(a) Analyses of diffraction intensities allow determination of surface corrugation functions, which reflect the electron charge-density contours^{1,2} and frequently reveal the geometry of the surface atoms directly.^{3,4} The relation $E(r) = \beta n_0(r)$ connects particle energy $E(r)$ and surface charge density $n_0(r)$ at the position r of the particle, β being a constant characteristic of the latter.¹

(b) The depth D of the attractive van der Waals potential, determined via selective adsorption resonances,⁵ can be used for surface composition diagnostics as demonstrated recently by Vidali *et al.* who confirmed that diamond (111) is covered with hydrogen.⁶

(c) The extreme sensitivity of He scattering to dilute adsorbates allows studies of surface migration as shown by Poelsema *et al.*⁷

(d) With use of time-of-flight techniques, surface phonon dispersions can be determined by inelastic He scattering.⁸

Surface investigations with Ne beams have been quite scarce up to now. Interestingly, there are more time-of-flight studies than diffractive investigations. This may be due to the prediction of Manninen *et al.*⁹ that $\beta\{\text{Ne}\} = 666 \text{ eV} \cdot a_0^3$, while $\beta\{\text{He}\} = 275 \text{ eV} \cdot a_0^3$; hence, Ne should scan smaller surface charge densities than He of the same energy, so that fewer corrugation details should be visible. The "ball rolling over the surface" picture³ leads to the same expectation in view of the larger size of Ne. On the other hand, the larger Ne mass causes enhanced inelastic scattering. However, Ne couples more strongly to bulk phonons than He,^{10,11} so that a differentiation between bulk and surface phonons is difficult with Ne, a stringent restriction on topic (d) above. Ne diffraction was hitherto only reported for surfaces with large corrugations like LiF(100)¹² and

Cu(117).¹³ Intensity analyses for LiF(100) gave similar corrugations for both He and Ne with a slightly smaller amplitude for Ne in accordance with the larger $\beta\{\text{Ne}\}$.⁹ A quantitative evaluation of Ne-diffraction data for the stepped Cu(117) surface was not possible because of the complicated shape and large amplitude of the corrugation.¹³

In the present paper we demonstrate that Ne scattering can contribute significantly to topics (a), (b), and (c). With regard to (a) and (b), we report for the first time Ne diffraction from relatively dense-packed metal surfaces, Ni(110) and Pd(110). Comparing with He diffraction in the same energy range, one surprisingly finds that Ne "sees" larger corrugations on these surfaces than He. Diffraction-intensity analyses yield similar corrugation shapes, but amplitudes which are much larger for Ne than for He, in contradiction to the reported β values⁹ and to the experimental result on LiF(100).¹² A possible reason for this unexpected effect is the stronger van der Waals interaction in the case of Ne, which might cause the classical turning points to lie closer to the surface-atom cores. This is supported by the fact that the repulsive potential of Ni(110) appears steeper for Ne than for He. Concerning (c) we observed that Ne is in general more sensitive to dilute adsorbates than He.

In our Ne-scattering experiments we used nozzle temperatures $T_N = 158\text{--}293 \text{ K}$, corresponding to beam energies $E_i = 34\text{--}63 \text{ meV}$ or wavelengths $\lambda_i = 0.347\text{--}0.255 \text{ \AA}$. The angle of incidence θ_i was varied between 25 and 60° . Typical in-plane ($\varphi = 0^\circ$) and out-of-plane Ne-diffraction scans for Ni(110) with $T_N = 190 \text{ K}$ ($\lambda_i = 0.316 \text{ \AA}$, $E_i = 40.9 \text{ meV}$), with the beam incident perpendicularly to the close-packed rows (y direction, $\gamma = 90^\circ$), and with the sample at $T_S = 100 \text{ K}$ are shown in Fig. 1. The intensity of the largest diffraction beams is $\sim 6\%$ of the incoming beam. The most significant and obvious feature, the pronounced rainbow near the beams ($0, \pm 2$), also

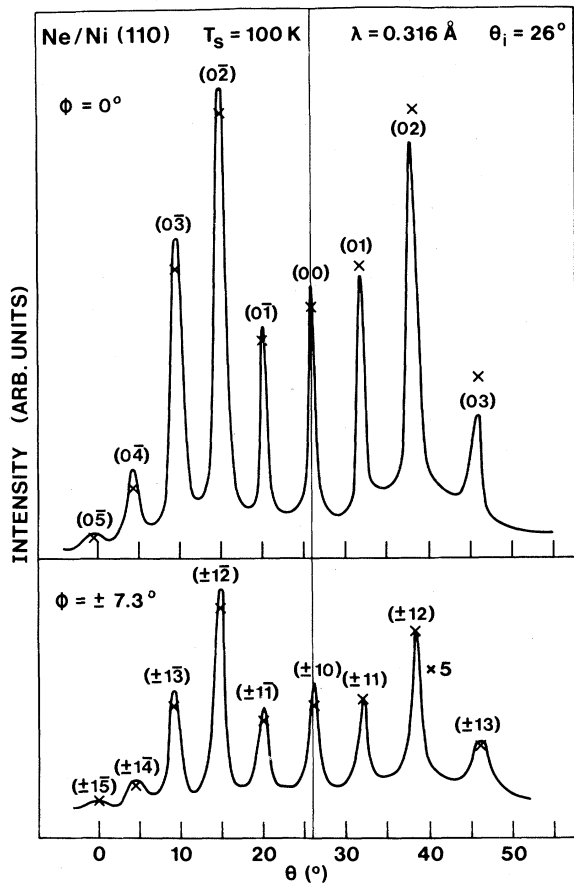


FIG. 1. Typical in-plane ($\phi = 0^\circ$) and out-of-plane Ne-diffraction traces for the clean Ni(110) surface. The beam is incident perpendicularly to the close-packed Ni rows. The best-fit peak intensities, with proper broadening of the beams due to the finite velocity spread taken into account, are indicated as crosses.

shows up in all other diffraction scans at different E_i and θ_i . Compared with He diffraction in the same energy range, where (0, ± 1) diffraction beams with only 10%–15% of the specular intensity were observed,³ the strong Ne rainbow immediately indicates a much larger corrugation amplitude than obtained with He. For quantitative evaluation, the hard-corrugated-wall (HCW) model¹⁴ and the simplest rectangular corrugation $z(x, y) = \{z(10) \cos(2\pi x/a_1) + z(01) \cos(2\pi y/a_2)\}/2$, with $a_2 = a_1\sqrt{2} = 3.52 \text{ \AA}$, were used. A good fit to all intensities is obtained with $z(10) = 0.033 \text{ \AA}$ and $z(01) = 0.174 \text{ \AA}$ [note that $z(10)$ determines the corrugation in the close-packed direction]. As a result of the larger polarizability of Ne, $\alpha\{\text{Ne}\} = 3.96 \times 10^{-25} \text{ cm}^3$, as compared to $\alpha\{\text{He}\} = 2.1 \times 10^{-25} \text{ cm}^3$, an almost twice as large value of D for Ne than for He can be expected.⁵ If one

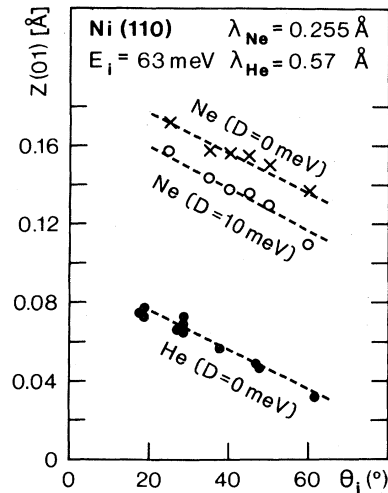


FIG. 2. Angular dependence of the $z(01)$ corrugation coefficients for Ni(110) obtained by HCW intensity analyses of diffraction spectra for room-temperature Ne and He beams. The different symbols indicate best-fit results; the dashed lines are guides to the eye. Refractive corrections for well depths $D \neq 0$ yield slightly smaller coefficients. The decrease of $z(01)$ with increasing θ_i is a measure of the softness of the repulsive potential and shows that the latter is softer for He than for Ne.

assumes that $D \approx 10 \text{ meV}$,¹⁵ and applies refractive corrections for E_i and θ_i ,³ slightly smaller parameters are obtained: $z(10) = 0.028 \text{ \AA}$ and $z(01) = 0.154 \text{ \AA}$ (the best-fit intensities are shown in Fig. 1). Debye-Waller corrections do not significantly affect these values, as an effective Debye temperature near the bulk value was found. Inelastic contributions to the diffraction intensities are small at low Ne energies (Fig. 1) and become increasingly important at higher E_i ; the errors made in evaluating data for higher E_i are nevertheless small as the inelastic intensities are proportional to the elastic ones¹⁶ and the calculations are all performed for intensities relative to specular.

Figure 2 shows the θ_i dependence of the HCW coefficient $z(01)$ for both He and Ne with use of room-temperature beams. The decrease of $z(01)$ with increasing θ_i is due to the softness of the repulsive potential.¹⁷ A quantitative evaluation of the softness parameter for Ne/Ni(110) was not possible as all methods for calculating diffraction intensities for soft potentials available to us^{18–20} failed to converge for the small Ne wavelengths and the relatively large corrugation. However, the fact that both curves in Fig. 2 have similar slopes indicates that the repulsive potential for

He is softer than for Ne. For larger corrugation amplitudes, the slope should be steeper with the same softness parameter as established by model calculations at longer wavelengths using a soft exponential potential.²⁰ On the basis of the HCW model the corrugation amplitudes can be reliably determined with use of data for small θ_i .¹⁷ Thus we can state that the corrugation amplitude observed for Ni(110) with Ne is about twice as large as for He, for which $z(10) = 0.015 - 0.02 \text{ \AA}$ and $z(01) = 0.075 \text{ \AA}$ in the same range of particle energies.¹⁷ Inclusion of higher-order Fourier coefficients improves the HCW fit of the Ne-diffraction intensities slightly; the resulting values are much smaller than the already small $z(10)$ showing that the shape of the corrugation is essentially the same for He and Ne.

Similar results were obtained for Pd(110) ($a_2 = 3.88 \text{ \AA}$). Figure 3 compares in-plane diffraction of Ne and He at $E_i = 63 \text{ meV}$ ($\gamma = 90^\circ$). The

most intense diffraction beam is $\sim 15\%$ of the incoming beam for He and only $\sim 1\%$ for Ne. Pronounced rainbows occur at $\sim 15^\circ$ for He and $\sim 40^\circ$ for Ne for all E_i and θ_i investigated; this again proves immediately that the corrugation for Ne is much larger than for He. HCW fits yield $z(01) = 0.21 \text{ \AA}$ for He with the measured depth $D = 8 \text{ meV}$ ²¹ taken into account and $z(01) = 0.425 \text{ \AA}$ for Ne with $D = 15 \text{ meV}$ as expected from $\alpha(\text{Ne})/\alpha(\text{He})$.⁵ It is remarkable that the simple sinusoidal corrugation yields a perfect description of the Ne rainbow as well as the correct sequence of intensities for the large number of diffraction beams [Fig. 3(a), dotted line]. A Ne background of presently unclear origin [Fig. 3(a), dash-dotted line] was observed for all E_i and θ_i . Out-of-plane diffraction data (not shown in Fig. 3) allowed determination of $z(10) \approx 0.04 \text{ \AA}$ for Ne and $\approx 0.02 \text{ \AA}$ for He.

We checked in two ways that the significantly larger corrugations observed with Ne are not due to Ne(1 \times 1) overlayers, which could form under the large effective pressure of the Ne beam: (i) With heating of the sample to 250 K the characteristic Ne-diffraction features remained; however, with an enhanced inelastic background. (ii) The relative intensities of the He-diffraction beams from both surfaces at 100 K did not change upon applying Ne background pressures equivalent to the Ne-beam pressures.

The observation that Ne "sees" appreciably larger corrugations than He for the metal surfaces investigated is in contradiction to Ref. 9 which predicts that the classical turning points for Ne should be farther away from the ion cores than for He, so that the lateral modulation of the electron density should be smaller. However, the existence of the attractive part of the potential modifies this simple picture: As pointed out recently by Liebsch and Harris²² for H₂ scattering from Ag(111),²³ a larger depth D of the attractive potential may cause the physisorption minimum and thus the classical turning points to lie closer to the surface-ion cores, so that a stronger corrugation is experienced. Although the effect should be stronger for H₂ than for Ne, as $\alpha\{\text{H}_2\}/\alpha\{\text{Ne}\} \approx 2$,⁵ it may cause the large corrugations observed with Ne for Ni(110) and Pd(110). Our finding that the repulsive potential of Ni(110) is steeper for Ne than for He supports this explanation. An alternative cause was proposed by Hill, Haller, and Celli,²⁴ who found that a strong lateral modulation of the attractive potential can give rise to enhanced diffraction. Our new Ne

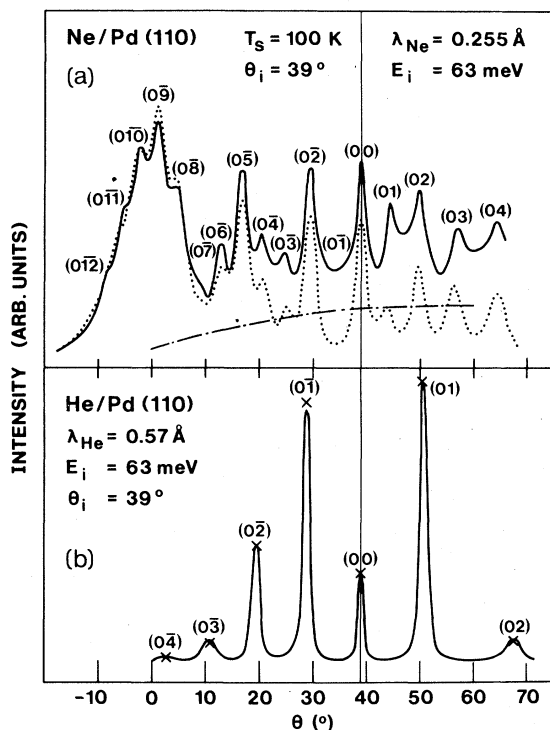


FIG. 3. Typical in-plane diffraction traces for the clean Pd(110) surface obtained (a) with Ne and (b) with He, with the beams incident perpendicularly to the close-packed rows. Best-fit intensities, with proper broadening of the beams due to the finite velocity spread taken into account, are shown as dotted lines in (a) and as crosses in (b). The background observed in the Ne scans is roughly indicated as a dash-dotted line in (a).

results should allow determination of the relative importance of these two possibilities.

Upon H adsorption on Ni(110), the initial rapid decrease of the specular intensity at low doses⁷ was found, for various scattering conditions, to proceed appreciably faster for Ne than for He, indicating that Ne is generally more sensitive to dilute adsorbates than He. A preliminary study of Ne diffraction from the 1×2 phase of H confirms the shape of the corrugation function found previously with He.³ However, in contrast to the clean metal surfaces and in agreement with the result on LiF¹² the corrugation amplitude is about the same as for He. It will be important to investigate whether the drastic enhancement of the corrugation seen with Ne is characteristic of clean metal surfaces.

In summary, we have shown that Ne scattering may be of great use in the first three areas of surface research discussed in the introduction: In regard to (a) and (b), with Ne appreciably larger corrugations are observed on dense metal surfaces than with He, so that more information on surface structure can be obtained as more details of the corrugation may become visible and more data on the particle-surface interaction potential such as bound-state energies, potential depths, and softness parameters may become measurable. In regard to (c), Ne is also more sensitive to dilute adsorbates and may therefore be used advantageously to detect small amounts of surface defects as well as to study surface diffusion.

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