

Scattering of He, Ne, Ar, H₂, and N₂ from Cu(110)

K. H. Rieder and W. Stocker

IBM Zurich Research Laboratory, 8803 Rüschlikon, Switzerland

(Received 9 October 1984)

Atomic-beam-scattering experiments on Cu(110) are reported for several gaseous species not reacting with the surface. Results are presented for the incoming beam impinging perpendicular to the close-packed metal rows. Diffraction features dominate for He, H₂, and Ne. The intensities of the He and H₂ diffraction beams relative to the specular are in very good agreement with the results of Salanon *et al.* [Surf. Sci. 127, 135 (1983)]. The hard-wall corrugation amplitude perpendicular to the close-packed rows obtained by fitting the He diffraction intensities shows a continuous increase with increasing He energy in contrast to the behavior we previously observed on Ni(110). Ne diffraction exhibits pronounced rainbows at $\sim 20^\circ$ from the specular showing that the corrugation amplitude seen with Ne is about twice as large as for He in complete analogy to the results previously reported on Ni(110) and Pd(110). Ar scattering shows rainbowlike features in the scattering plane with the rainbow angle smaller than in the case of Ne. N₂ scattering exhibits only broad in-plane scattering around the specular, whereas unexpected pronounced out-of-plane structure is observed.

I. INTRODUCTION

In the last few years, He diffraction has proven to be a very valuable tool for surface structural investigations.¹⁻⁴ Besides being an absolutely nondestructive method owing to the very small particle energies of 20–200 meV, which also allows investigation of very subtle adsorption phases,²⁻⁴ it has the advantages of probing the topmost layer of the surface only, being sensitive to light adsorbates on heavy substrates^{2,3} and—last but certainly not least—the diffraction intensity analyses do not require any model assumptions on the surface structures.³ Intensity analyses yield the so-called corrugation function, which is a replica of the electron-density contour corresponding to the normal component of the incoming particle energy.^{5,6} Frequently, the corrugation function immediately shows the geometry of the surface atoms relative to one another, and also contains important information on surface bonding.^{4,7} Quite recently, pronounced Ne diffraction has been observed for clean metal surfaces⁸ adding new prospects for surface structural research with atomic beams.

The present work pursues several purposes: First, it provides the first comparison of He and H₂ diffraction data on the same metal surface obtained independently in two different laboratories (the other being represented by Refs. 9–12). At the same time, the He diffraction results underscore that the energy dependence of the corrugation of the (110) surface of Cu is different from that of Ni(110).^{13,14} Secondly, our Ne diffraction results on Cu(110) corroborate our previous findings on the (110) surfaces of Pd and Ni,⁸ that with Ne about twice the corrugation amplitude as with He is seen. Finally, our investigations of the scattering of the heavier particles Ar and N₂ yield new and unexpected features.

II. EXPERIMENTAL

The Cu(110) specimen disk of 6-mm diameter and 1-mm thickness was prepared from a large Cu single-crystal boule. The crystal was oriented to $\pm 0.3^\circ$ and then cut

with a wire saw. The (110) surface was first mechanically polished with successively finer grades of diamond paste. An electropolishing procedure using an aqueous solution of phosphoric and sulfuric acid followed. After several Ne-bombardment and annealing cycles in ultrahigh vacuum, the low-energy electron diffraction pattern was sharp and clear and an Auger electron spectroscopic analysis with our retarding field analyzer showed no traces of impurities.

Atomic- and molecular-beam-scattering experiments with the sample at 105 K were performed for all five species with the nozzle at room temperature and pressures behind our 10 μm nozzle of 5 bar for H₂ and 20–30 bar for all other gases. For He, H₂, and Ne, for which diffraction predominates, cooled and heated nozzles were also used.² Diffraction-intensity analyses were performed in the framework of the hard corrugated-wall model¹ mainly with the eikonal formula,¹⁵ which is well justified since the corrugation amplitude of the clean Cu(110) surface is sufficiently small for all three species. All analyses could be performed with the simple one-dimensional corrugation function,

$$\zeta(x) = \frac{1}{2} \zeta(01) \cos \left(\frac{2\pi}{a} x \right), \quad (1)$$

with $a = 3.61 \text{ \AA}$ being the distance between the close-packed metal rows and x denoting the crystallographic [001] direction. This shows that the shape of the corrugation (which contains the main crystallographic information as it visualizes the arrangement of the surface atoms) is the same for all three species.

III. RESULTS AND DISCUSSION

A. He diffraction

A typical diffraction scan obtained with the room-temperature He beam is shown in Fig. 1(a). The diffraction peaks are appreciably stronger relative to the specular

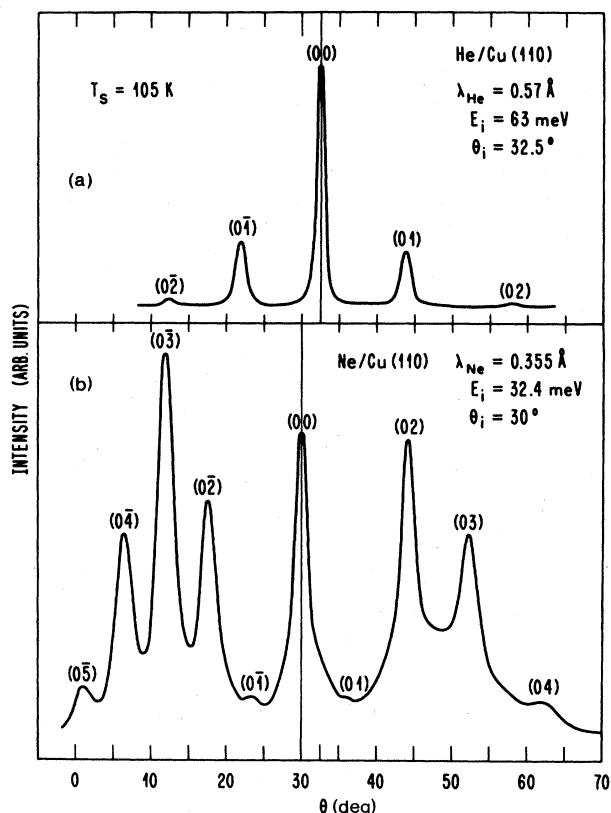


FIG. 1. (a) Typical He diffraction scan obtained with the room-temperature beam for Cu(110). The beam is incident perpendicular to the close-packed rows. (b) Typical Ne diffraction scan obtained with a cooled beam (nozzle temperature 152 K). Note that in contrast to the He pattern, pronounced rainbow structure is observed at about 20° from the specular. This observation shows directly that the corrugation amplitude obtained with Ne is appreciably (about a factor of 2) larger than that obtained with He.

than in the case of Ni(110). The relative intensities compare very favorably with the data published by Salanon *et al.*⁹ as shown in Fig. 2. It must be emphasized that, again in agreement with Salanon *et al.*,⁹ we did not observe any out-of-plane diffraction beams; this is in strong contrast to Ni and Pd(110), and shows that the corrugation along the close-packed metal rows is much smaller for Cu than for Ni and Pd. Figure 3 compares the hard corrugated-wall (HCW) amplitudes $\zeta(01)$ of Cu(110) and Ni(110) obtained with the room-temperature beam data for different angles of incidence. Refraction effects owing to the attractive part of the particle-surface interaction potential have been taken into account in the best-fit calculations for Cu [the approximate potential depth $D \approx 6$ meV was determined by Perreau and Lapujoulade¹¹ from selective adsorption resonances on Cu(113), Cu(115), and Cu(117) surfaces]. As expected from the good agreement in the intensities measured by Salanon *et al.*⁹ and ourselves, the agreement of the HCW parameters is also ex-

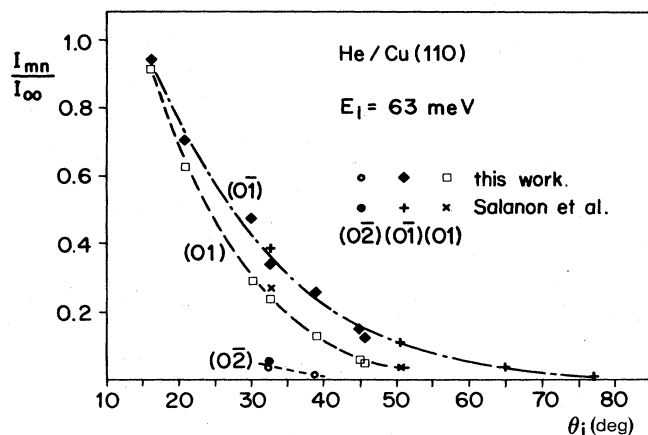


FIG. 2. Comparison of He diffraction intensities relative to the specular between our work and that of Salanon *et al.* (Ref. 9) shows very good agreement between both laboratories.

cellent. The decrease of $\zeta(01)$ with increasing angle of incidence constitutes a coarse measure of the softness of the He-surface potential. Since the decrease between 20° and 50° is slightly less than a factor of two for both Ni and Cu the softness is similar for both metals. Indeed, for Ni(110) the softness parameter was determined to be $\sim 2.6 \text{ \AA}^{-1}$,¹⁴ and a similar value^{10,11} was obtained for Cu.

According to the theoretical work of Esbjerg and Norskov⁵ and Harris and Liebsch,⁶ the corrugation function is a replica of the surface-charge-density contour corresponding to the energy of the momentum normal to the surface $E_n = E_i \cos^2 \theta_i$.^{13,14} Thus, in general increasing corrugation amplitudes with increasing energy are to be expected. Figure 4 shows that this is indeed the case for Cu(110); for the angle of incidence fixed at 33°, the He-

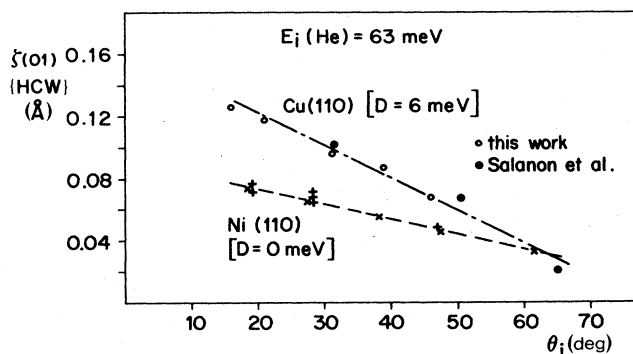


FIG. 3. Hard-wall corrugation amplitude perpendicular to the close-packed metal rows as a function of angle of incidence for fixed He energy. The results for Cu(110) are again in good agreement with the data of Salanon *et al.* (Ref. 9). Analogous data for Ni(110) (Ref. 13) are also shown. The decrease of the HCW amplitude with increasing angle of incidence constitutes a measure of the potential softness; the fact that the relative decrease is about the same for both metals shows that the softness is very similar in both materials.

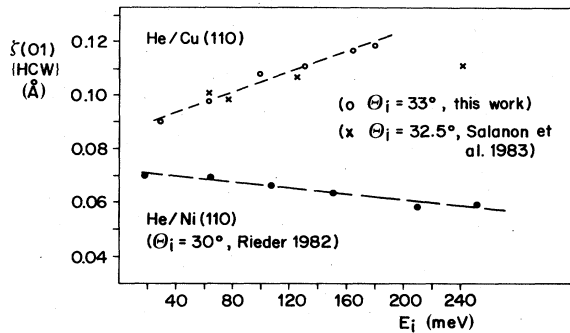


FIG. 4. Hard-wall corrugation amplitude perpendicular to the close-packed metal rows as a function of energy for a fixed angle of incidence. In contrast to Ni(110), the data for Cu(110) show the expected increase of $\zeta(01)$ with increasing energy.

corrugation amplitude increases by about 30% between 30 and 180 meV. Our results in this kind of measurement again agree very nicely with the ones of Salanon *et al.*,⁹ when we omit their point at 240 meV (Fig. 4). The good agreement of our results on Cu(110) with those of the collaborators of Refs. 9–12 also suggests that our measurements on Ni(110) (Refs. 13 and 14) may be trusted. Therefore the observation previously reported (also reproduced in Fig. 4) that the Ni-corrugation amplitude $\zeta(01)$ obtained for the same scattering configuration shows a slight downward trend has to be regarded as further confirmed. It should be noted in this context that the softness of the potential influences the Ni(110) data reproduced in Fig. 4 insofar as it gives rise to a more rapid decrease of the amplitude the larger the angle of incidence becomes. At small angles, where the softness effects have the smallest influence, the corrugation amplitude of Ni(110) remains roughly constant (for an extended discussion of these effects see Ref. 13). Applying this observation to Cu(110), we may expect that for smaller angles of incidence the increase of $\zeta(01)$ as a function of energy may become slightly larger. This was indeed observed for $\theta_i = 23^\circ$, where we found an increase of about 40%, from ~ 0.95 to ~ 0.135 Å, between 20 and 180 meV. In view of the good agreement of our intensity data with those of the collaborators of Refs. 9–12, we did not perform any data analysis beyond the hard corrugated-wall model as the results obtained with more realistic models (like the corrugated Morse potential) on the basis of our data would also be very similar to those of Salanon *et al.*,⁹ Perreau and Lapujoulade,¹⁰ and Harris and Liebsch.¹⁶

B. H₂ diffraction

Figure 5 shows a typical diffraction scan obtained with the room-temperature beam of H₂. A hard corrugated-wall analysis using Eq. (1) and incorporating refractive corrections with the rather large potential depth of 22 meV as determined by Perreau and Lapujoulade¹¹ (see also Ref. 17) for the hard-wall corrugation amplitude $\zeta(01)$ as a function of angle of incidence, yields the behavior shown in Fig. 6. The agreement with the data of Lapu-

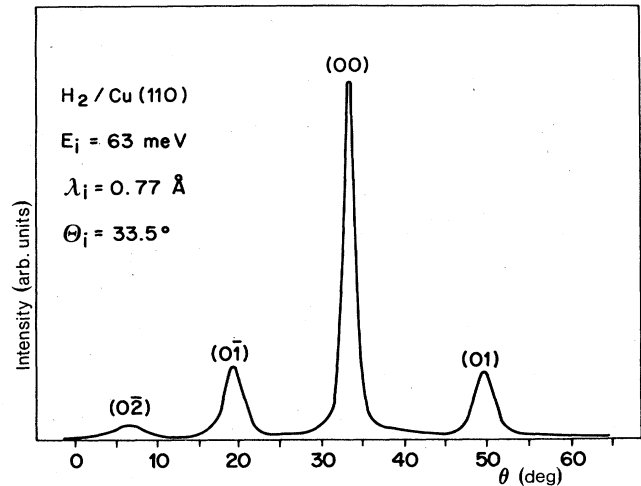


FIG. 5. Typical H₂ diffraction scan obtained with the room-temperature beam for Cu(110). The beam is incident perpendicular to the close-packed rows.

joulade and Perreau¹² is satisfactory although not as good as for the case of He diffraction. It is worth noting that owing to the large value of the potential depth the corrugation amplitudes are appreciably smaller than the values obtained when the refractory correction is neglected.

C. Ne diffraction

According to the theoretical predictions of Puska *et al.*¹⁸ the proportionality constant relating surface charge density and normal energy of the incoming particle should be larger for Ne than for He.⁸ In the simple picture of the Esbjerg-Norskov approach,⁵ which neglects the influence of the attractive van der Waals part of the interaction potential, this means that the classical turning points of the Ne atoms should be farther away from the surface ion cores than in the case of He. Thus it was expected [and indeed observed for LiF(100) (Ref. 19)] that the corrugation amplitudes for Ne should be slightly

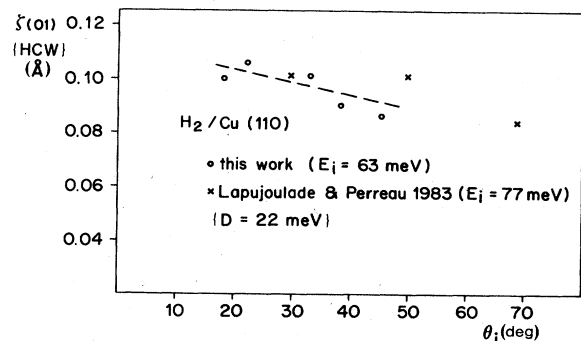


FIG. 6. Comparison of HCW amplitudes obtained from H₂ diffraction data on Cu(110) in our laboratory and that of Ref. 12.

smaller than for He. It was therefore a great surprise, when we recently succeeded in observing pronounced diffraction effects with Ne on the clean (110) surfaces of Ni and Pd, that the hard-wall model framework yielded corrugation amplitudes about twice as large as for He.

One of the main aims of the present investigation was therefore to provide further confirmation of this effect on another clean metal surface. A typical result of our Ne-scattering experiments on Cu(110) is compared in Fig. 1 with a diffraction result obtained with He. All Ne-diffraction patterns exhibit a pronounced rainbow at an angular distance of $\sim 20^\circ$ from the specular. In the case of an incoming Ne beam cooled to 152 K as shown in Fig. 1(b), the rainbow occurs near to the (03) diffraction beam. With the room-temperature Ne beam, the rainbow is clearly at the (04) diffraction beam as shown in Fig. 7(a). In the case of He diffraction with the room-temperature beam [Fig. 1(a)], the first-order beams are much smaller than the specular. Only at the highest He energies investigated (~ 180 meV) do the first-order beams exceed the specular in intensity, and even then the higher-order beams are appreciably smaller. Comparison of the diffraction spectra in Fig. 1 immediately shows that, as evidenced by the strong rainbow scattering, the corrugation

amplitude for Ne is appreciably larger than that for He. This is in full analogy to the previous observations on Ni(110) and Pd(110).⁸

A more quantitative evaluation of the data was performed by fitting the intensities experimentally observed to the hard-wall corrugation of Eq. (1). According to Ref. 17, the depth of the attractive potential is expected to be twice as large for Ne as for He, as the ratio of polarizabilities of Ne and He is roughly a factor of two.^{8,17} Therefore, refractive corrections with a potential depth of 12 meV (Ref. 10) were included. Very good fits were obtained for all intensities measured, proving that the shape of the corrugations observed with He and Ne is similar. For small angles of incidence (25° – 35°), where softness effects play only a negligible role, the corrugation amplitudes $\zeta(01)$ were found to lie between 0.21 and 0.23 Å in the energy range between 30 and 64 meV. Thus, the corrugation amplitude is again almost twice as large as in the case of He, where according to Fig. 3 (extrapolation to $\theta_i = 0^\circ$) we find a value of $\zeta(01) \sim 0.13$ Å to be representative for $E_i = 63$ meV. It should be noted that this value is in satisfactory agreement with the more involved analyses of Salanon *et al.*⁹ based on the corrugated Morse potential.

Another observation concerns the softness of the potential. As discussed in Sec. III A, the angular decrease of the corrugation amplitude for a given energy provides a coarse measure of the softness parameter. Using the room-temperature beam data obtained with Ne, we observe a decrease from 0.23 to 0.17 Å between $\theta_i = 25^\circ$ and 50° . This is roughly a decrease of 25%, and a similar value was also observed for Ne/Ni(110).⁸ In contrast to this result, the decrease in the case of He is much stronger, namely $\sim 50\%$, both for Cu(110) (see Fig. 3) and Ni(110).^{8,13} According to model calculations performed with the soft exponential potential,^{19,20} we can conclude from this finding that the repulsive potential experienced by the Ne atoms is about twice as steep as that felt by the He atoms.

It is very remarkable that although Ne obviously penetrates deeper into the sea of surface electrons and is thus able to observe more details of the corrugation shape, in the case of Cu(110) we did not observe any out-of-plane diffraction, as was also the case with He (Sec. III A). The Ne result is proving even more strikingly that the smearing-out effect of the free electrons is much more effective along the close-packed metal rows in Cu than in Ni and in Pd, where with He weak and with Ne appreciable, out-of-plane diffraction peaks were found.^{13,8}

As mentioned at the beginning of this section, the result that, on clean metal surfaces, with Ne much larger corrugation amplitudes are observed than with He, is in contradiction to the expectations based on the simple Esbjerg-Norskov picture;⁵ according to which Ne should scan lower densities and thus see smaller corrugations than He. In this simplified approach, the influence of the attractive part of the potential is omitted. As was pointed out in a theoretical study of Liebsch and Harris,²¹ an increased depth of the physisorption potential may help the incoming particles to penetrate closer to the surface ion cores so that they would scan corrugations corresponding

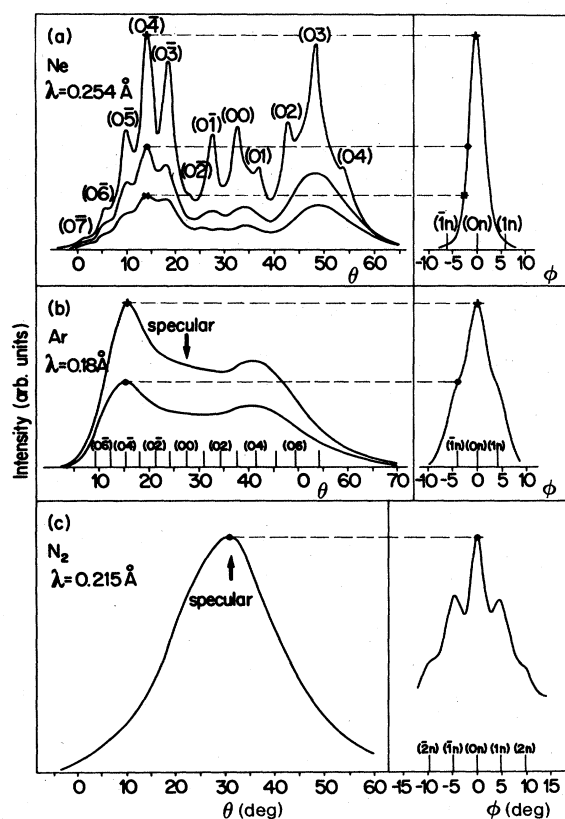


FIG. 7. In-plane and out-of-plane scans for scattering of room-temperature beams of: (a) Ne, (b) Ar, and (c) N₂ from Cu(110). The vertical lines in (a), (b), and (c) indicate the positions of the diffraction beams expected according to the respective room-temperature beam wavelengths.

to effectively larger densities than expected according to the Esbjerg-Norskov proportionality constant. It appears reasonable to argue that such an effect would be more pronounced at soft surfaces such as metals, in which the decay of the electron density perpendicular to the surface is slower than on harder surfaces such as ionic materials. The influence of the potential depth, which should be about twice as deep for Ne as for He,¹⁷ may therefore explain on the one hand the strongly different results obtained with He and Ne on the "soft" (110) surfaces of the metals Ni, Cu and Pd, and on the other hand the very similar results obtained on the rather "hard" (100) surface of LiF.²² Measurements with He and Ne on the same system should thus allow convenient and quick checks as to whether the surface is soft or hard. The effect of the attractive potential should be even more pronounced with Ar because of its much larger polarizability as compared to Ne,¹⁷ but, as discussed in the following section, this does not seem to be the case for Cu(110).

D. Scattering of Ar and N₂

In order to investigate whether diffraction effects would still be observable with beams of heavier particles, we performed further scattering experiments on Cu(110) with Ar and N₂. In Fig. 7, typical results obtained with room-temperature beams of Ar [Fig. 7(b)] and N₂ [Fig. 7(c)] are contrasted to the Cu(110) diffraction spectrum obtained with a Ne room-temperature beam [Fig. 7(a)].

With both Ar and N₂, the in-plane scans showed broad intensity distributions in which individual diffraction peaks are not discernible. For the case of Ar, the possible diffraction angles are indicated by vertical lines. Owing to the larger mass of the Ar atoms, inelastic contributions are expected to be much stronger than in the case of Ne, where they are already significantly stronger than in the case of He scattering. It is unclear at present whether the expected strong inelastic contributions smear out the regions between diffraction peaks so effectively that they become invisible, or whether diffraction effects are indeed vanishingly small in the case of Ar scattering. Nevertheless, with Ar, pronounced peak structures in the in-plane intensity distributions are observed reminiscent of the rainbow-scattering features found with Ne. The Ar rainbow angle as measured from the specular is $\sim 13^\circ$, significantly smaller than in the case of Ne, where it was found to be $\sim 20^\circ$. We present two arguments which favor the presence of diffraction in the Ar case: First, the locations of the rainbow maxima agree rather well with the (expected) diffraction angles of the (0 $\bar{4}$) and (04) beams, and secondly the rainbow maxima do not shift with increasing surface temperature.

Consequently, should the rainbow actually stem from diffraction effects, we are faced with the remarkable phenomenon that Ne "sees" the largest corrugation amplitude in the series of noble gases He, Ne, and Ar. As the polarizability of Ar is about four times larger than that of Ne,¹⁷ which implies a potential depth approximately four times larger,¹⁷ we should—in the spirit of the discussion in Sec. III C—expect the Ar atoms to scan the electron density even closer to the surface ion cores than the Ne,

unless the Esbjerg-Norskov proportionality constant for Ar is much larger than for Ne so that the latter effect outweighs the former.

Another interesting phenomenon is observed with Ar in ϕ scans perpendicular to the scattering planes. The results are exhibited on the right side of Fig. 7, wherein the ϕ location of the θ scan is indicated by the dashed horizontal lines. In the case of Ne, we observe a rapidly decreasing intensity behavior with the same width as characteristic for the in-plane diffraction beams. No out-of-plane diffraction beams are observed, as discussed in Sec. III C, indicating a complete lack of corrugation parallel to the close-packed Cu rows. With Ar, however, we observe a pronounced shoulder at $\phi \simeq 4^\circ$ which coincides with the ϕ location of the out-of-plane ($\pm 1n$) beams.

The latter effect appears especially interesting in the light of the results obtained with N₂ scattering, Fig. 7(c). In this case, in-plane we observe only a broad intensity distribution peaking in the specular direction. Out of plane, we observe an even more pronounced structure than in the case of the Ar-scattering experiments: There are clear peaks at $\phi = \pm 4.8^\circ$, and pronounced shoulders at $\phi \simeq 9^\circ$; the coincidence with the expected locations of the ($\pm 1n$) and ($\pm 2n$) diffraction peaks, again indicated as vertical bars, is striking.

At present, we cannot offer any explanation for the effects observed with Ar and N₂. The fact that with He and Ne on Cu(110) no out-of-plane diffraction is found, whereas with Ar and N₂, out-of-plane diffractionlike features seem to be present, is entirely puzzling. Therefore, we are of the opinion that the results presented here are sufficiently interesting to pursue them further experimentally, preferably by using high-resolution time-of-flight apparatus²³ to allow for clear separation of elastic diffraction intensities from inelastic scattering contributions. In the case of N₂ scattering, one has furthermore to remember that during the collision with the surface rotational degrees of freedom of the molecules are also certainly involved. These could be partly responsible for the peculiar scattering distributions reported here, and state selective detection of the scattered particles²⁴ would therefore be valuable in this case.

IV. CONCLUSIONS

We have found very satisfactory agreement between the He and H₂ diffraction data on Cu(110) obtained with our atomic beam-scattering apparatus and the results reported earlier in Refs. 9–12, on the same surface. The strong dependence of the hard-wall corrugation amplitude on the angle of incidence of the incoming beam shows that the surface softness is similar to the case of Ni(110). The corrugation amplitude perpendicular to the close-packed rows as measured with the He beam at small angles of incidence (where softness effects do not play a role) is found to amount to ~ 0.13 Å for a beam energy of 63 meV. This value is larger than the corresponding value obtained for Ni(110) (0.08 Å) and smaller than that for Pd(110) (0.22 Å). This sequence correlates well with the increasing lattice constants of Ni, Cu, and Pd. In this context, it is also very remarkable that for Cu(110) the corrugation

along the close-packed rows is vanishingly small in contrast to the observations on the (110) surfaces of Ni and Pd. Again in contrast to Ni(110), where the corrugation perpendicular to the close-packed metal rows is found to be practically independent of the incoming energy, the same parameter exhibits for Cu(110) a continuous increase with increasing energy.

In the case of Ne scattering from Cu(110), we observe—as for Ni(110) and Pd(110)—that diffraction dominates and that the corrugation amplitude perpendicular to the close-packed rows is much larger (about a factor of 2), than for He diffraction. The corrugation amplitude in the close-packed direction is again found to be vanishingly small. The repulsive potential for Ne is approximately twice as steep as for He, which has the important consequence that the hard-corrugated-wall model works much better for Ne than for He in the case of clean metal surfaces. The reason for this effect may lie in the influence of the attractive part of the potential which is twice as deep for Ne than for He and may therefore cause the turning points for Ne to lie closer to the surface ion cores

than for He.

Puzzling results were obtained with Ar and N₂ scattering: Rainbowlike features in-plane are observed for Ar, although no well-resolved diffraction peaks are discernible. Surprising in view of the Ne results is the observation that the Ar rainbow angle is smaller than for Ne! Pronounced out-of-plane structure of unknown origin is observed near the angle of first-order out-of-plane diffraction beams. N₂ scattering shows only broad in-plane distribution around specular direction but pronounced out-of-plane structure at first- and second-order diffraction angles. These observations are not understood at all at present and clearly require (and deserve!) further more detailed investigations.

Note added. In a very recent paper [J. Phys. (Paris) **45**, 1373 (1984)], B. Salanon applied the corrugated Morse potential to analyze Ne diffraction data from Cu(110), and arrived at the result that the softness parameter for Ne is 1.8 times longer than for He, in excellent agreement with our present conclusions.

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