theoretical and experimental investigations are required to clarify the inconsistency among these presumably related experiments.

This work was supported in part by the Research Corporation and by the U. S. Department of Energy. We thank R. L. Fagaly for several helpful discussions on the experimental techniques.

[1]D. H. Dye, S. A. Campbell, G. W. Crabtree, J. B. Ketterson, N. B. Sandesara, and J. J. Vuillemin, Phys. Rev. B 23, 462 (1981).
[2]B. M. Boerstoel, J. J. Zwart, and J. Hansen, Physica (Utrecht) 54, 442 (1971).
[3]For example, G. W. Crabtree, D. H. Dye, D. P. Karim, and J. B. Ketterson, J. Magn. Magn. Mater. 11, 236 (1979).
[4]N. F. Berk and J. R. Schrieffer, Phys. Rev. Lett. 17, 433 (1966).
[5]S. Doniach and S. Engelsberg, Phys. Rev. Lett. 17, 750 (1966).
[6]See, for example, H. Ramm, P. Pedroni, J. R. Thompson, and H. Meyer, J. Low Temp. Phys. 2, 539 (1970).
[7]R. J. Trainor, M. B. Brodsky, and H. V. Culbert, Phys. Rev. Lett. 34, 1019 (1975).
[8]W. F. Brinkman and S. Engelsberg, Phys. Rev. 169, 417 (1968).
[9]M. T. Béal-Monod, S.-K. Ma, and D. R. Fredkin, Phys. Rev. Lett. 20, 929 (1968).
[10]P. Hertel, J. Appel, and D. Fay, Phys. Rev. B 22, 534 (1980).
[11]J. R. Schrieffer, Phys. Rev. Lett. 19, 644 (1967).
[12]D. Fay and J. Appel, Phys. Rev. B 16, 2325 (1977).
[13]N. B. Sandesara and J. J. Vuillemin, Metall. Trans. 8B, 693 (1977).
[14]R. L. Fagaly and R. G. Bohn, Rev. Sci. Instrum. 48, 1502 (1977).
[15]D. L. Martin, Can. J. Phys. 47, 1253 (1969).
[16]G. Choteau, R. Fourneaux, R. Tournier, and P. Lederer, Phys. Rev. Lett. 21, 1082 (1968).
[17]T. D. Cheung and J. S. Kouvel, to be published.
[18]O. K. Anderson, Phys. Rev. B 2, 893 (1970).
[19]T. Jarlborg and A. J. Freeman, to be published.
[20]K. Ikeda and K. A. Gschneidner, Jr., Phys. Rev. Lett. 45, 1341 (1980).
[21]K. Ikeda and K. A. Gschneidner, Jr., to be published.
[22]K. Ikeda and K. A. Gschneidner, Jr., and O. D. McMasters, to be published.
[23]F. A. Muller, R. Gersdorf, and L. W. Roeland, Phys. Lett. 31A, 424 (1970).

Momentum Transfer Cutoff in the Scattering of Neon Atoms from a Nickel (111) Surface

B. Feuerbacher
Space Science Department of the European Space Agency, European Space Research and Technology Centre, 2200-AG Noordwijk, The Netherlands

and

R. F. Willis
Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, United Kingdom

(Received 20 April 1981)

Angle-resolved time-of-flight measurements of inelastic scattering of Ne atoms from a Ni(111) surface show an effective interaction radius increased with respect to the Ne hard-sphere radius, such that the Ne atoms impact against five Ni atoms. This limits the interaction with acoustic surface waves to some critical cutoff wave vector, K_c. Larger energy transfers are accompanied by vanishing momentum transfer as assumed in a cubes model.

PACS numbers: 68.20.+t, 79.20.Rf

A gas atom (or molecule) colliding with a solid exchanges energy and momentum with the surface. The incident particle may be scattered elastically, inelastically, or it may become trapped at the surface, depending on the amount of energy transferred. The exchange of energy and momentum is therefore of fundamental importance for all processes involving dynamic gas-surface interactions. It is usually described phenomenologically in terms of an energy accommodation coefficient,[1,2] while the underlying microscopic processes are still a matter of current debate.

Measurements of He scattering from alkali halide surfaces[3] have shown that the interaction with single Rayleigh surface phonons is the predominant energy-transfer process between light

gas atoms and ionic crystal surfaces. A stronger contribution from bulk phonons was suggested[4] for the interaction of Ne atoms with the same crystal. Considerable differences are expected in the scattering behavior from metal surfaces due to the role played by the conduction electrons. The continuum of low-energy electron-hole pair excitations may participate in the scattering process either directly (by electron-hole pair excitation) or indirectly (by limiting the phonon lifetimes). The spatial distribution of the conduction electrons has been shown to strongly reduce diffraction effects by smearing the potential variations between the ion cores.[5] Recent results for inelastic He scattering from a Cu(100) surface[6] show spectra that appear distinctly different from those obtained on alkali halides and which have been interpreted in terms of single-phonon interaction with both bulk and surface excitations.

The present results demonstrate a different effect occurring for the interaction of a relatively heavy atom (Ne) with a close-packed metal surface [Ni(111)]. Here the blanket of conduction electrons at the surface acts to distribute the interaction forces over several metal atoms, thus effectively increasing the gas-atom-interaction radius. This leads to a continuumlike scattering dominated by multiphonon effects, rather than the stricly atomic pair-wise interaction via single Rayleigh phonons observed on insulators,[3] and to a cutoff wave vector for the excitation of acoustic surface waves.

In the system studied here, viz. Ne interacting with a Ni(111) surface, the well depth of the interaction potential is small (~10 meV) compared with the energy of the incident thermal gas atom (64.5 meV), so the energy exchange can be conveniently studied by scattering methods. The measurement consists of scattering a near-monochromatic, collimated beam of atoms from the surface and observing the change in velocity distribution as a function of the scattering angle. The results are analyzed in terms of energy and momentum transfer between gas atom and surface. The measurement system, described elsewhere,[7] uses a room-temperature pulsed-gas beam generated in a fast switching valve source[8] with an opening time of 10 to 20 μsec and a velocity monochromaticity of about 7%. A fixed angle of $2\theta_s = 135°$ was maintained between beam source and detector while rotation of the sample by an angle $\Delta\theta$ changed incidence angle $\theta_i = \theta_s - \Delta\theta$ and observation angle $\theta_f = \theta_s + \Delta\theta$ simultaneously, with the scattering plane parallel to the [112] azimuth. The scattered-gas pulses were detected after a flight distance of 905 mm in a commercial nude ion gauge, with use of a fast amplifier and analog-to-digital conversion for storage in a multichannel system.

A set of typical time-of-flight spectra of 64.5-meV neon atoms scattered from a room-temperature Ni(111) surface is shown in Fig. 1. The center curve marked 0° represents specular scattering at $\theta_i = \theta_s = 67.5°$. The lower spectra marked by negative sample angles $\Delta\theta$ correspond to increased angles of incidence, and scattering closer to the surface normal. The evaluation in terms of energy and momentum transfer is illustrated in Fig. 2. Here the uppermost panel shows a time-of-flight spectrum for $\Delta\theta = 14°$ (full line) together with the specular spectrum (dashed line). The flight time can be converted directly into energy transfer ΔE with use of the independently measured beam velocity and the known flight distances. This is shown in the center panel of Fig. 2, where negative energy values correspond to an energy loss of the scattered atoms. Assuming conservation of the parallel momentum component in the scattering event, the momentum transfer parallel to the surface ΔK can be derived for

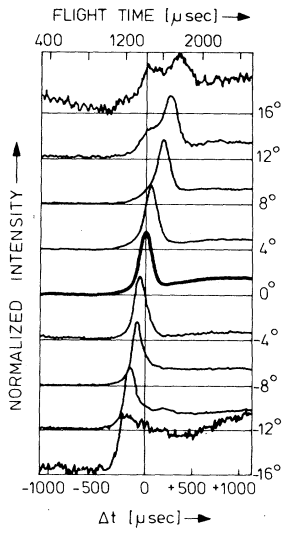

FIG. 1. Time-of-flight spectra of 64.5-meV Ne scattered from a room-temperature Ni(111) surface, for different angles of sample rotation as given along the right-hand side. The heavy curve (0°, specular scattering at $\theta_i = 67.5°$) is about 50% broader than the incident velocity distribution. The lower scale gives the time difference to the specular peak arrival time.

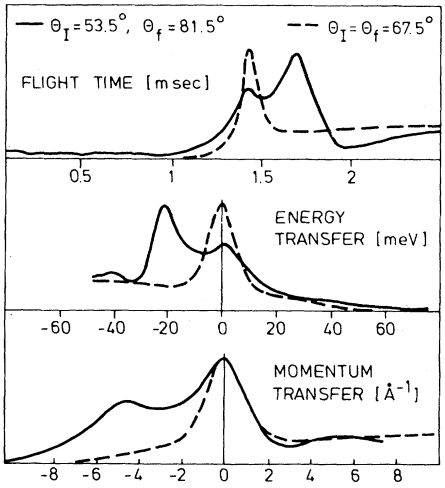

FIG. 2. Ne scattered from Ni(111) at $\Delta\theta = 14°$ compared with specular scattering (dashed lines). The top panel gives the measured time-of-flight spectra. The lower panels show the same spectra transformed into energy space or momentum space as indicated.

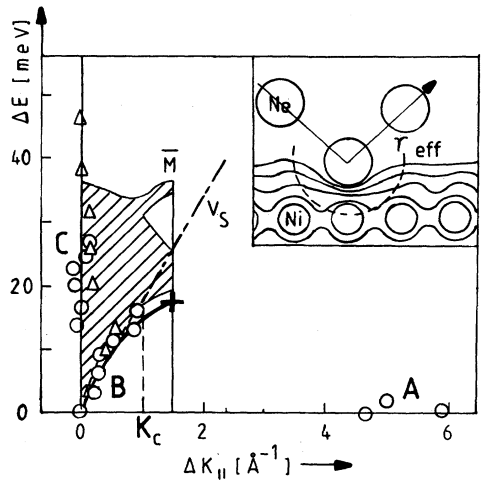

FIG. 3. Absolute values of energy vs parallel momentum transfer in Ne-Ni(111) scattering as derived from time-of-flight spectra. Circles and triangles correspond to energy loss and gain, respectively. The shaded area gives the phonon spectrum along the $\bar{\Gamma}\bar{M}$ line (Ref. 10). The plus represents the energy of the Rayleigh phonon at $\bar{M}$ as observed by electron-energy-loss spectroscopy (Ref. 12). Inset: Schematic of electron density distribution near the Ni surface during the impact of a Ne atom.

planar scattering[9]

$$\frac{\Delta K}{K_i} = 1 - \left(1 + \frac{\Delta E}{E_i}\right)^{1/2} \frac{\sin\theta_f}{\sin\theta_i}, \qquad (1)$$

where K_i and E_i are the incident parallel wave-vector component and beam energy, respectively. Using this transformation, the same data have been plotted as a function of momentum transfer in the lower panel of Fig. 2, with negative values of ΔK being parallel to the incident wave vector.

These peak positions have been monitored for a variety of measured angles and values for ΔE and ΔK determined. The results are plotted in Fig. 3 modulus their absolute values such that energy gains or losses, as well as momentum gains or losses, appear in the first ΔE vs ΔK quadrant. Also shown is the phonon band structure calculated[10] along the $\bar{\Gamma}\bar{M}$ symmetry line on the Ni(111) face which has been normalized to fit the bulk phonon spectrum,[11] and a value for the surface acoustic wave recently reported at point $\bar{M}$ from electron-energy-loss spectroscopy studies[12] (marked by the plus in Fig. 3). The observations fall into three distinctly different categories of behavior, *A*, *B*, and *C* (Fig. 3).

Group *A*, characterized by large momentum transfer but with vanishingly small energy transfer, can be attributed to elastic, but *incoherent* scattering from nonperiodic local surface irregularities. A similar feature has been observed in earlier measurements on ionic crystals[3] and metals.[6] Group *B* points show a linear relationship between ΔE and ΔK which corresponds, within the accuracy of the measurements, to the surface-sound-wave velocity dispersion in the continuum limit (dash-dotted line, Ref. 13) or equally, with the Rayleigh-wave phonon curve (full curve), up to a critical value of transferred parallel momentum $\Delta K = K_c$. Then, higher energy transfers occur extending well beyond the maximum single-phonon energies available such that points in group *C* (Fig. 3) are indicative of a process in which the parallel momentum transfer ΔK remains effectively zero. This is in complete contrast to the recent observations reported for helium scattering from LiF[3] or Cu,[6] where single-phonon scattering was shown to predominate over the complete range of energy and momentum transfer observed.

The interpretation of the measurement summarized in Fig. 3 proceeds according to a model shown by the diagram in the inset in Fig. 3. Here an effective interaction radius r_{eff} is assigned to the impinging atom, which is larger than the Ne hard-sphere radius. Acoustic surface waves can only be excited for wavelengths larger than this value r_{eff}, equivalent to a specific cutoff wave vector $K_c = \pi/r_{\mathrm{eff}}$. This corresponds to the

linear dispersion found for group B, where the observed value of K_c leads to an effective interaction radius of $r_{\text{eff}} \cong 3$ Å. The model therefore suggests that the incident Ne atoms impact against approximately five Ni atoms in the close-packed Ni(111) surface, in good agreement with a value of approximately four surface atoms deduced from the angular and velocity distributions of argon atoms scattered from a polycrystalline tungsten surface.[14] As the magnitude of the energy transfer increases beyond the critical value, $\Delta E = V_s K_c$, coupling to acoustic surface waves is no longer possible. The surface acts as an assemblage of flat cubes of face size related to r_{eff}, which reflect the impacting atoms without parallel momentum transfer and move normal to the surface with velocities corresponding to the surface temperature. Only the normal velocity component of the scattered atom is changed on impact, registering an increase (decrease) depending on whether the cube face is moving up (down) at the moment of collision; thus neon atom energy gains (losses) are found for scattering angles towards (away from) the surface normal.

The picture emerging for large energy transfers coincides fully with the hard-cubes model[15] that has been used widely and successfully for the interpretation of the lobular structure of heavy atom scattering from a metal surface. The present results in fact justify the main assumption of the cubes models (viz. $\Delta K = 0$) and mark its range of validity against the limit of single-phonon models. The crucial parameter is the effective interaction radius r_{eff}, which depends on the mass ratio, the impact energy, and the electronic screening properties. If r_{eff} is smaller than the surface unit cell dimension, single-phonon effects are expected to dominate. This is, e.g., the case for the scattering of He from a Ni(111) surface, where no momentum cutoff is observed.[16] In contrast, for heavy atom scattering one expects[17] $r_{\text{eff}} \gg a$, so the contribution of surface-wave excitation will be negligible and a cubes model will describe the interaction satisfactorily.

The authors are indebted to Bill Gadzuk and Bengt Lundqvist for their valuable comments. They are particularly grateful to John Harris for introducing the hard-cube model into the present interpretation. This work was supported in part by the Royal Society (London).

[1]R. M. Logan, in *Solid State Surface Sciences*, edited by M. Green (Marcel Dekker, New York, 1973), Vol. 3, p. 2.

[2]F. O. Goodman and H. Y. Wachman, *Dynamics of Gas-Surface Scattering* (Academic, New York, 1976).

[3]G. Brusdeylins, R. B. Doak, and J. P. Toennies, Phys. Rev. Lett. 44, 1417 (1980), and 46, 437 (1981).

[4]L. Mattera, C. Salvo, S. Terreni, F. Tommasini, and U. Valbusa, to be published.

[5]G. Boato, P. Cantini, R. Tatarek, J. Phys. F 6, L237 (1976).

[6]B. F. Mason and B. R. Williams, Phys. Rev. Lett. 46, 1138 (1981).

[7]W. Allison and B. Feuerbacher, Phys. Rev. Lett. 45, 2040 (1980).

[8]M. R. Adriaens, W. Allison, and B. Feuerbacher, to be published.

[9]G. Benedek, Phys. Rev. Lett. 35, 234 (1975).

[10]R. E. Allen, G. P. Alldredge, and R. W. de Wette, Phys. Rev. B 4, 1661 (1971).

[11]R. J. Birgeneau, J. Cordes, G. Dolling, and A. D. B. Woods, Phys. Rev. 136, A1359 (1964).

[12]H. Ibach and D. Bruchmann, Phys. Rev. Lett. 44, 36 (1980).

[13]G. W. Farnell, in *Physical Acoustics*, edited by W. P. Mason and R. N. Thurston (Academic, New York, 1970), Vol. VI, p. 109.

[14]E. K. Grimmelman, J. C. Tully, and M. J. Cardillo, J. Chem. Phys. 72, 1039 (1980).

[15]K. C. Janda, J. E. Hurst, C. A. Becker, J. P. Cowin, D. J. Auerbach, and L. Wharton, J. Chem. Phys. 72, 2403 (1980).

[16]B. Feuerbacher, unpublished.

[17]See, e.g., J. L. Beeby, in *Dynamic Aspects of Surface Physics*, edited by F. O. Goodman (Editrice Compositori, Bologna, 1974), p. 751.