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Critical Cone in Phonon-Induced Desorption of Helium

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A flash desorption technique is used to show that helium atoms desorbing from a hot surface are emitted within a narrow cone about the normal direction. The emission angle and the energy of the atoms are highly correlated, which results in a "rainbow" in the desorption spectrum. These effects are discussed in terms of single-particle conservation conditions for energy and momentum transfer between phonons and atoms.

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Phonons in a solid substrate can interact with atoms on a surface by causing displacements which modulate the local surface potential. An adsorbed atom initially in a bound state can be desorbed by this perturbation. A quantum theory of this process of phonon-induced desorption was first considered by Lennard-Jones and co-workers,[1] but several more sophisticated treatments have recently appeared.[2-6] Using these theories, one can express the momentum distribution of the desorbed atoms in terms of a perturbation series expansion involving the phonon distribution and the surface-adatom interaction. First-order perturbation theory yields a value for the probability of a 1-phonon desorption process of the form $|M|^2/\omega$, where ω is the phonon frequency and $|M|^2$ is a squared matrix element which depends only on the initial and final states of the atom; the probability of multiphonon desorption processes is related to higher orders of the perturbation which are much more complicated and difficult to compute.[6]

Although the 1-phonon approximation has been useful in explaining atomic beam scattering results,[7] it is difficult to test the simple desorption theories decisively using only the available data on the desorption rate and the energy distribution. In contrast, the complete three-dimensional (3D) distribution of the atomic momentum provides a much more sensitive probe of the desorption process. Previous measurements of the angular distribution of desorption[8,9] have involved systems at such high temperatures and with such strong adatom-substrate interactions that the 1-phonon perturbation theories are not applicable. Helium, with a binding energy an order of magnitude smaller than most other adsorbates, provides a good test case for theories based on the weak coupling approximation which suggest that at low temperatures 1-phonon processes should be the dominant desorption mechanism. Quantitative comparison with the published theories is difficult, however, because of uncertainties in the parameters which determine $|M|^2$; moreover, the most detailed calculations assume that the adatom is localized to a site on the substrate, which is probably not an appropriate model for helium on a metallic surface.[10] Despite these

difficulties, much of the physics of the 1-phonon picture can be understood in terms of simple kinematic constraints which are independent of the precise form of $|M|^2$. The purpose of this Letter is to examine some of the implications of 1-phonon kinematics and to compare the predictions with experimental results on the angular and energy distribution of helium desorbed from Nichrome.

Assuming that the surface-atom interaction potential is translationally invariant in the plane of the surface, the kinematical constraints on the 1-phonon desorption process are given by the conservation of energy and parallel momentum:

$$\omega = c|\vec{k}| = p^2/2m + E_b, \tag{1}$$

$$\vec{k}_{\parallel} = \vec{p}_{\parallel}, \tag{2}$$

where c is the phonon phase velocity, $\vec{k}$ is the phonon k vector, $\vec{p}$ is the momentum of the free atom, and E_b is the binding energy of the atom to the surface. The rate $\dot{N}(E, \vec{p}_{\parallel})$ at which atoms with kinetic energy E and parallel momentum $\vec{p}_{\parallel}$ are desorbed is

$$\dot{N}(E, \vec{p}_{\parallel}) = \int_{\omega > c|\vec{k}|} \frac{|M|^2}{\omega} c f(\omega, T_s)\, \delta(E - \omega + E_b)\, \delta(\vec{k}_{\parallel} - \vec{p}_{\parallel})\, d\omega\, d^2k_{\parallel} \tag{3}$$

where $f(\omega, T_s)$ is the phonon distribution function, T_s is the substrate temperature, and the inequality which defines the region of integration ensures that only real incoming phonons are included.

The velocity and angular distribution of the desorbed atoms $\dot{n}(v, \Omega)$ is easily obtained from Eq. (3) by changing variables and performing the integration[11]:

$$\dot{n}(v, \Omega) = \begin{cases} \dfrac{|M|^2 v^3 \exp[-(E_b + mv^2/2)/T]}{E_b + mv^2/2} \cos\theta, & \text{if } mv^2/2 + E_b > mcv \sin\theta, \\ 0, & \text{otherwise}, \end{cases} \tag{4}$$

where Ω is the solid angle and the approximation $f(\omega, T) \cong e^{-\omega/T}$ has been substituted. Although the explicit dependence on angle has the $\cos\theta$ form expected from an equilibrium distribution, the inequality introduces a velocity-dependent cutoff in angle. The qualitative implications of Eqs. (1) and (2) and the inequality constraint are graphically illustrated in Fig. 1, which shows curves of constant energy in momentum space for atoms and phonons. For a given incident phonon, the momentum vector $\vec{p}$ of the desorbed atom can be found by a simple geometric construction which shows that the atoms are emitted into a cone with a critical half-angle of approximately 10°. Because the atom energy is a quadratic function of momentum while the phonon energy is linear in momentum, the exact value of the critical angle is energy dependent. This results in a rainbowlike effect in which low-energy atoms can escape from the surface at larger angles than can high-energy atoms, which are emitted almost normal to the surface, as shown in Fig. 1(b).

The experimental apparatus used to test these implications of energy and parallel-momentum conservation is schematically shown in the inset of Fig. 2. A sapphire crystal which has a Nichrome heater film (0.4×0.2 mm²) on one surface is mounted on a vertical turntable in a vacuum can. The turntable is attached to a shaft which is supported by ball bearings and is connected to a similar shaft at the top of the Dewar with a pulley and belt arrangement. A tin superconducting transition bolometer (0.02×0.02 mm²) fabricated on the end of a sapphire rod is used to detect the helium atoms which are desorbed from the heater when a voltage pulse (typically 2 V×100 nsec) is applied. The distance between the heater and bolometer was 1.1 mm with the heater precisely positioned over the stationary axis of the turntable. The angle between the heater surface normal and the bolometer was measured with a capacitance technique. The ambient temperature was 2.09 K, the helium-gas pressure in the cell was 3×10^{-7} Torr, and the atom binding energy $E_b = 30$ K, as measured by techniques described elsewhere.[12,13]

The bolometer signal for various angular positions is shown in Fig. 2. The signal intensity is a rapidly decreasing function of angle, reaching half its normal-incidence value at 15°. In addition, the average arrival time of the atoms increases with increasing angle, or equivalently, the average energy decreases with angle, in accordance with the qualitative results suggested

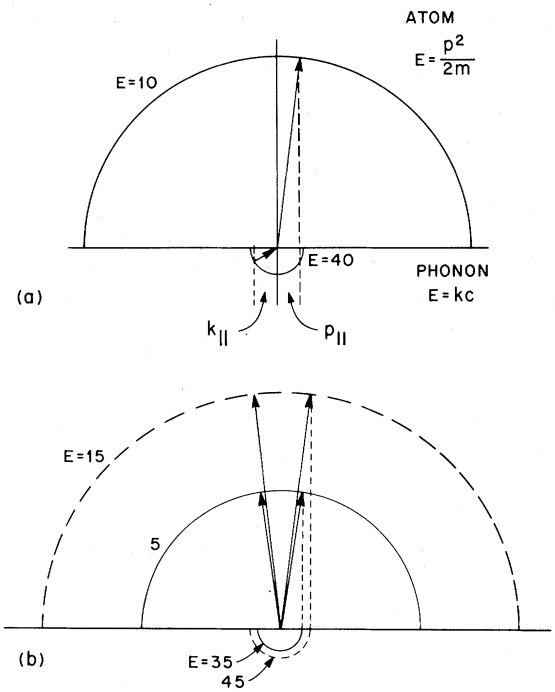

FIG. 1. Curves of constant energy in momentum space for atoms above the interface, and phonons below, drawn to scale with $c = 3\times10^5$ cm/sec and $E_b = 30$ K (all energies in kelvins). (a) Geometric construction which shows that parallel-momentum conservation leads to atoms emitted almost normal to the interface. (b) The angular width of the atomic critical cone depends on the energy, with high-energy atoms emitted into a narrower cone. This results in an atomic rainbow in the desorption spectrum.

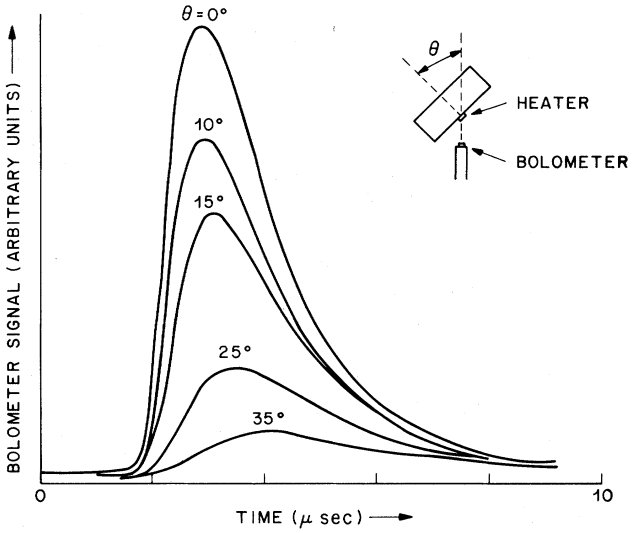

FIG. 2. Bolometer signal as a function of time for several values of θ. $T_s = 8$ K.

by Fig. 1.

To compare the data with the quantitative predictions of 1-phonon kinematics, it is necessary to compute the expected bolometer signal, $S(t, \theta)$. Since the bolometer is a linear detector of energy flux, the signal is proportional to the product of the rate at which atoms arrive at the bolometer and the total energy carried by each atom:

$$S(t, \theta) = A\dot{n}(v, \theta)(mv^2/2 + E_b)v^2, \quad (5)$$

where A is a proportionality constant, l is the distance between the heater and bolometer, and t is the arrival time at the detector with $v = l/t$. For a given value of the substrate temperature T_s, which can be computed from the heater power by acoustic-mismatch theory, Eqs. (4) and (5) determine the form of the expected bolometer signal due to 1-phonon desorption. For $\theta = 0$, the time dependence of $S(t, \theta)$ agrees with the data of Fig. 2 quite well, but Eq. (5) predicts a more abrupt decrease in the atom flux with increasing θ than is observed in the experiment. For example, at $\theta = 15°$, 1-phonon kinematics suggests that only atoms with energy less than 1.7 K or greater than 517 K would be able to reach the bolometer, and the predicted signal would be much too small to measure.

The observed angular distribution, although strongly peaked in the forward direction, is approximately twice as wide as Eq. (3) would predict and the atoms which leave the surface at angles larger than 10° have anomalously low velocity. The fact that atoms are observed to reach the detector with more parallel momentum than a phonon alone could impart could be due to several effects. The possibility that the critical cone of Fig. 1 is broadened by surface roughness was checked by calculating the signal expected from a surface with a Gaussian distribution of normal vectors. Although a surface with an angular rms deviation of 25° would widen the angular distribution sufficiently to agree with the observations, the correlation between angle and velocity is destroyed; i.e., there would be no rainbow from a rough surface.

Another effect which tends to broaden the cone is the initial thermal parallel momentum of the atoms on the surface. The geometric construction which leads to the critical cone of Fig. 1 assumes that the adatom is stationary, i.e., at $T = 0$, but in a more realistic description the helium atoms can be modeled as a 2D ideal gas at the ambient temperature T_a with a Maxwellian

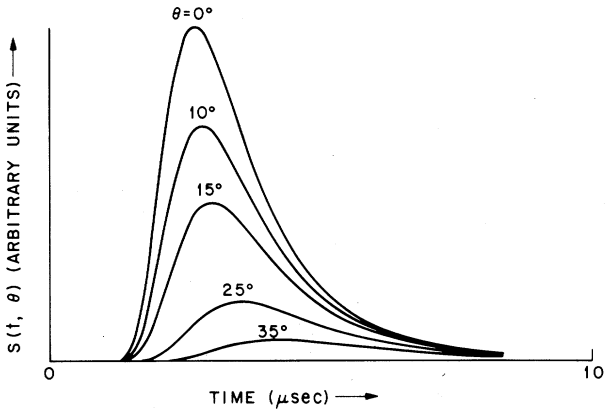

FIG. 3. Computed curves of $S(t,\theta)$, the expected bolometer signal, for the same experimental conditions as in Fig. 2, including the initial thermal atomic momentum.

distribution of momentum in the plane. Although the parallel component of thermal energy of the atom is small compared to the phonon energy ω, the momentum of an atom at 2 K is approximately equal to the momentum of the phonons which cause desorption, as can be seen in Fig. 1; so this effect could potentially account for the factor-of-2 increase in the width of the cone. The results of a detailed calculation of the bolometer signal $S(t,\theta)$ which includes the initial energy and parallel momentum of the atoms in the conservation conditions Eqs. (1) and (2) is shown in Fig. 3. The convolution of the simple 1-phonon signal with the initial thermal momentum distribution accounts for the angular width as well as the velocity-angle correlation in the observed signal very well. The theoretical curves are quite sensitive to the temperature which characterizes the Maxwellian distribution of parallel momentum. If the temperature of the helium film was raised by as little as 1 K by phonons which were absorbed and thermalized, the resulting angular distribution would be noticeably broader than is observed.

In summary, I have presented the first combined measurements of the energy and angular distribution of phonon-desorbed helium. Helium atoms are desorbed into a cone centered about the normal direction with a half-angle of approximately 15°. There is a strong correlation between the energy of the desorbed atom and the angle at which it leaves the substrate, producing a rainbowlike effect in the momentum distribution. The average energy of the atoms with $\theta = 30°$ is approximately half that of the atoms which desorb in the normal direction. Remarkably, the features in the angular distribution are not superimposed on a broad background; there is no observable signal for $\theta > 45°$. These effects are qualitatively consistent with the kinematics of a phonon-atom interaction assuming conservation of energy and parallel momentum. If the initial thermal distribution of the atom momentum is taken into account, the quantitative agreement between the model and the data is very good.[14] This agreement suggests that the 1-phonon picture may accurately describe the desorption of helium. It is possible that the large ratio of atom momentum to phonon momentum may lead to very anisotropic desorption distributions even for multiphonon processes, but to evaluate the contribution of higher-order processes or to put limits on them will not be possible until more detailed calculations on the expected angular and velocity distributions are available.

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[14]The calculations described in Refs. 2 and 5 also lead to strongly peaked distributions, but in these models, parallel momentum is not conserved and the angular width is governed by the transverse variation of the binding potential around the adsorption site. For reasonable binding-potential parameters, the parallel mo-

mentum of the atom due to the zero-point energy associated with the transverse localization has a distribution which is very similar to a particle in a 2D ideal gas at a temperature of a few kelvins. Thus, in these models, the critical cone is broadened by the ground-state momentum of the localized adatoms, but the resulting desorption distribution is similar to the results of Fig. 3.

Defect Structures below the Surface in Metals Investigated by Monoenergetic Positrons

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A new compact system has been developed to produce monoenergetic positrons of variable energy. The energy range available at present is between 150 eV and 28 keV, so that depth profiles can be measured. This positron beam is being used to investigate defects and defect structures close to the surface in metals and alloys. The technique of Doppler broadening of the annihilation radiation is applied for these measurements.

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The positron annihilation technique has been used for many years for the investigation of various defects and defect configurations in metals and alloys.[1] The positron is a very sensitive probe in particular for vacancylike defects and vacancy agglomerates. Since changes in the positron annihilation characteristics can be observed already for vacancy concentration as low as 10^{-7} this method is especially useful for low defect concentrations. Because of this high sensitivity, positrons can be used to study the vacancy agglomeration to voids in metals, which is also of great technological interest with respect to materials used in high-flux fission and fusion reactors. Positrons emitted from a radioactive source have an energy distribution with maximum energies between 0.5 and 1.5 MeV for various isotopes. Thus the penetration depth of the positrons into the sample to be investigated is of the order of 100 μm. Therefore in order to obtain results which are characteristic for the status of the sample, the concentration and the distribution of the defects has to be homogeneous over the total range of the positrons. For monoenergetic positrons of variable energy the damaged region can be relatively thin and close to the surface, since the penetration of the positrons can be adjusted according to their energy. By varying the energy of the positrons, different damage profiles in various depths from the surface can be sampled. This opens the possibility of investigating materials irradiated by low-energy light ions (H, He) and heavy ions (self-ions) by positron annihilation techniques. The first measurements on neutron- and helium-irradiated metals are reported here.

Positrons from a 25-mCi ^{22}Na source are thermalized in a 10-μm gold foil covered with MgO (positron converter).[2] A fraction of about 10^{-4} of the positrons are remitted from the converter foil with energies of approximately 1-2 eV. By electrical and magnetic lenses, these positrons are focused to form a positron beam of about 6 mm diameter and are then deflected 180° in a magnetic field. At present any energy between 150 eV and 28 keV can be obtained. Therefore the penetration profile of the positrons can be varied from a few times 10^{-10} to 10^{-6} m. The vacuum in the target chamber is 3×10^{-10} Torr and a lock system is installed to facilitate changing specimens while maintaining ultrahigh-vacuum conditions. The intensity of the positrons at the target is 10^5 per second. A tungsten collimator is placed in such a way that the high-purity Ge detector used for the Doppler-broadened 511-keV γ rays only measures annihilations emanating from the target itself.

A positron-annihilation Doppler-broadening spectrum provides momentum-distribution information about the electrons with which the positron annihilates. In well-annealed metal samples the positron annihilates while in a delocalized state in the essentially defect-free lattice. In irradiated samples positrons can be trapped at various defects and annihilate then from a localized state, and the annihilation characteristics give information about the nature of the defects. Results on neutron- and helium-irradiated copper specimens are shown in Fig. 1. All positron annihilation measurements were performed at

