ANISOTROPY OF THE ENERGY SPECTRA OF SLOW ALKALI-METAL IONS SCATTERED FROM SINGLE-CRYSTAL TARGETS

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An anisotropy of the energy spectra of slow K^+, Rb^+, and Cs^+ ions scattered from incandescent single-crystal targets (Mo and W) has been observed experimentally. The dependence of the maximum energy of the scattered ions W_m on the angle of incidence χ of the primary ions with respect to various crystal planes for a fixed scattering angle ψ has been investigated. The maximum of the curve $W_m(\chi)$ shifts to larger values of χ with decrease of ψ. These maxima are due to scattering of the primary ions either from crystal planes with a maximum atomic packing density (Cs^+, Rb^+) resulting from unpaired interaction between the ion and lattice atoms or along the most closely packed directions on the target surface (K^+) resulting from multiple pair collisions. The relative role of these collisions depends mainly on the "size" of the primary ion. Schematized calculations are presented which confirm the interpretation of the results obtained.

At present there are very few experimental data on the scattering of ions with energies $U \lesssim 1-2$ keV from the surface of a single crystal[1] and no information at all on the shape of the energy spectra of these ions and its dependence on the orientation of the primary ion beam with respect to the crystal lattice. Nevertheless, this information is of interest in the theory of interaction of ions with the surface of a solid. Furthermore, data of this type can make possible the determination of the shape, so far little studied, of the repulsive potential between heteronuclear systems of atoms with unclosed electron shells.[2,3]

In the present work we have investigated the energy spectra of Cs^+, Rb^+, and K^+ ions scattered by monocrystalline targets (tungsten and molybdenum) at a given angle ψ for different angles of incidence χ of the primary beam with respect to the target surface. Particular attention was paid to study of the dependence of the maximum energy of the scattered ions on the angle χ, since this dependence characterizes the interaction of the particle with the target in the simplest form.[4] Ideal experimental conditions are guaranteed by the fact that the scattering coefficient of the alkali-metal ions is quite large,[5] the targets were used in the incandescent state (1500—1600°K), and the primary ion energies were so low (100—260 eV) that they could not produce a noticeable sputtering of the target material in the form of ions.[6]

The apparatus, the experimental technique, and the method of determining the maximum energy of the scattered ions W_m have been basically described earlier[4] (the measurements utilized a method of supplying voltages based on circuit 2). The targets were cut in the form of strips approximately $1.5 \times 0.15 \times 0.015$ cm from single crystals of tungsten or molybdenum. Before cutting, the crystals were electrolytically polished and their orientation determined with a GD-1 goniometer in which 10—15 faces were examined. In cutting the targets we endeavored to have their long axis coincide with previously determined directions having low crystallographic indices. However, we did not always succeed in this. To avoid the effect of mechanical working on the structure of the target surface, the specimens were finished to the required thickness by electrolytically etching at least 0.02 cm of material from the surface.[7] After preparation of the target, a final determination was made of the crystal orientation.

Contrary to our expectations[1,8], we did not observe a noticeable dependence of the total yield of the fast[4] scattered ion group on crystallographic direction. On the contrary, the shape of the energy spectra of the scattered ions, which are generally similar to the spectra for polycrystalline targets,[9] and the value of W_m turned out to be strongly dependent on the orientation of the primary beam with respect to the crystal lattice of the target.

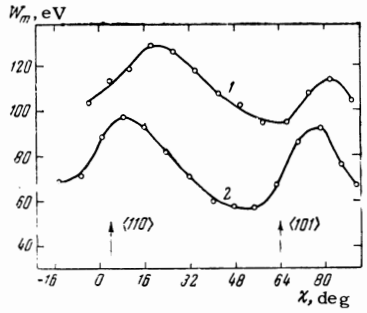

FIG. 1. Maximum energy of scattered Cs^+ ions as a function of the angle of incidence of the primary ions, for rotation of a tungsten target around the <$\bar{1}11$> direction (U = 200 eV): 1 – ψ = 90°, 2 – ψ = 126°.

Figure 1 shows the experimentally obtained variation of W_m with angle of incidence χ for two values of the scattering angle ψ of Cs^+ ions from a tungsten target whose axis of rotation coincided with the $\langle\bar{1}11\rangle$ direction. Here the $\langle 110\rangle$ and $\langle 101\rangle$ directions lay in the plane of rotation S of the normal to the target macrosurface. The angle χ here and subsequently will be considered positive if the external normal to the surface and the momentum of the scattered particle at some point of the target's surface lie on the same side of the primary ion momentum at this point. The range of variation of the angle χ was 126°. Naturally, the accuracy of the measurements for the extreme positions of the target (target surface parallel to the primary beam or to the analyzer axis[4]) was lower than for intermediate values of χ. Together with the anisotropy in W_m observed in Fig. 1, our attention is drawn to the displacement of the peaks in the $W_m(\chi)$ curves towards higher values of χ with decreasing scattering angle.

The data shown in Fig. 2 were obtained for the case when the axis of rotation of the target was inclined from the $\langle 001\rangle$ direction by an angle Δ = 17°, and the $\langle 010\rangle$, $\langle 110\rangle$, and $\langle 100\rangle$ directions

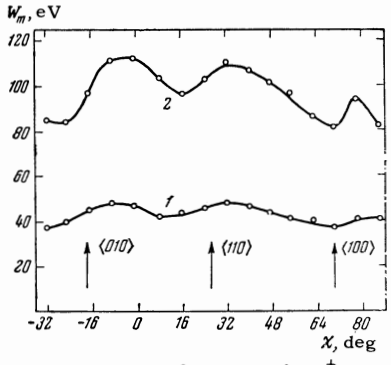

FIG. 2. Maximum energy of scattered Cs^+ ions as a function of the angle of incidence of the primary ions for rotation of a tungsten target around an axis close to <001> (ψ = 126°): 1 – U = 100 eV, 2 – U = 260 eV.

formed angles of 9, −3, and −15° with the plane S, respectively. The observed reduction in the height of the peak for deviation of a certain crystallographic direction from the plane S (cf. $\langle 010\rangle$ and $\langle 100\rangle$) is in agreement with the interpretation suggested below. It follows from the data shown that the shape of the function $W_m(\chi)$ is almost independent of the primary ion energy. A similar behavior was observed also for rotation of the target around a $\langle\bar{1}11\rangle$ axis.

We can attempt to interpret the results in the following way. As before, let us assume that the scattering involves the strong interaction of the primary ion simultaneously with a group of lattice atoms. Let a system consisting of four lattice atoms possess a four-fold symmetry axis C_4 (Fig. 3) and let the primary ion move with a velocity $\mathbf{v}_0$ in one of the planes of symmetry of this system not passing through the centers of the atoms. In this case there are two pairs of atoms, in each of which both atoms (M_1, M_2 and correspondingly M_3 and M_4) receive the same momentum. Let this momentum be MV_4 for the first pair and kMV_4 for the second pair (here and subsequently M is the mass of the lattice atom, m is the mass of the primary ion). We will designate by ϕ the angle between the directions of $\mathbf{v}_0$ and C_4; the sign of the angle ϕ is determined in the same way as the sign of the angle χ, as a function of the positions of the vectors $\mathbf{v}_0$ and $\mathbf{v}_4$ ($\mathbf{v}_4$ is the velocity of the ion after scattering) with respect to the axis C_4. For given values of v_0 and ϕ the value of k depends on the relative location of the vector $\mathbf{v}_0$ and the center of mass of the lattice atoms being discussed. The probability that k has some particular value is determined by quantities similar to the differential cross sections for pair collisions. The latter must depend very strongly on the form of the scattering potential in the system. However, for simplicity we will discuss a collision in the approximation of absolutely hard elastic spheres. Here a simultan-

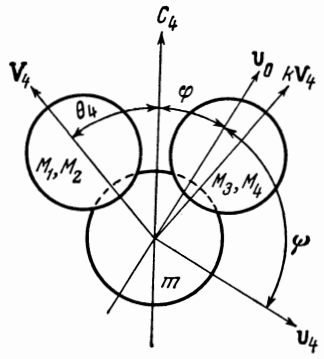

FIG. 3. Drawing illustrating the collision of an ion with four atoms.

eous collision of the ion with the four lattice atoms will be possible only in the case when the center of the ion at the moment of collision lies on the axis C_4 and the quantity k enters into the equations as a parameter.

This approach does not allow us to find the probability of the collision discussed but it permits us to analyze the important characteristics of the scattering. Specifically, let θ_4 be the angle included, at the moment of collision, between the axis C_4 and each of the lines joining the center of the ion and one of the atoms. Then the laws of conservation of energy and momentum have the following form:

$$\frac{mv_0^2}{2} = \frac{mv_4^2}{2} + 2\frac{MV_4^2}{2}(1+k^2),$$
$$mv_0 \cos\varphi = mv_4 \cos(\varphi+\psi) + 2MV_4(k+1)\cos\theta_4,$$
$$mv_0 \sin\varphi = mv_4 \sin(\varphi+\psi) + \sqrt{2}MV_4(k-1)\sin\theta_4. \quad (1)$$

Eliminating V_4 and ϕ from Eq. (1), it is easy to show that

$$v_4 = v_0 \frac{\cos\psi \pm [(M_{eff}/m)^2 - \sin^2\psi]^{1/2}}{1+M_{eff}/m}, \quad (2)$$

$$M_{eff} = 2M\left\{\frac{(k+1)^2}{k^2+1}\cos^2\theta_4 + \frac{1}{2}\frac{(k-1)^2}{k^2+1}\sin^2\theta_4\right\}. \quad (3)$$

It follows from formula (3) that the assumption made earlier that the maximum value of M_{eff} corresponds to a "quasisymmetric collision" (k = 1)[4] is actually correct for $\theta_4 \leq 54.5°$. The quantity k is not determined experimentally but is related to the angles ϕ and ψ. Therefore, if we eliminate V_4 and k from the Eqs. (1), the ion velocity after the collision is given by the following relation:

$$v_4 = v_0 [2\cos^2\theta_4 \sin(\varphi+\psi)\sin\varphi + \sin^2\theta_4 \cos(\varphi+\psi)\cos\varphi \pm \alpha_4 \sin 2\theta_4] \times [\beta_4 + \sin^2\theta_4 + \gamma_4 \sin^2(\varphi+\psi)]^{-1} \quad (4)$$

where

$$\alpha_4 = {}^1/_2[\mu_4\beta_4 - 2\sin^2\psi + \mu_4\gamma_4\{\sin^2(\varphi+\psi) - \sin^2\varphi\}]^{1/2},$$
$$\beta_4 = \mu_4\cos^2\theta_4\sin^2\theta_4, \quad \gamma_4 = 2\cos^2\theta_4 - \sin^2\theta_4,$$
$$\mu_4 = 4M/m.$$

For $\psi = \pi$ this formula takes on an already well known form,[4] and for $\theta_4 = 0$ it becomes the formula for pair collisions. It is not difficult to show that for $\mu_3 = 3M/m$ formula (4) is applicable also to the case of collision of an ion with three lattice atoms having a threefold symmetry axis C_3. In the case where there is no symmetry the expression for v_3 becomes quite complicated. However, even in this case it is possible to use formula (4) with a sufficient degree of accuracy.

In the expression for v only the angle θ is unknown. Under actual conditions the value of this angle is determined by the slowing down of the ion and lies between zero and a value θ_{max} determined from the condition of greatest penetration of the electron shells of the ion and the lattice atoms. The true value of the angle θ depends strongly on the shape of the potential for repulsion of the electron shells of the ion and the lattice atoms. Therefore we will consider θ as a parameter. Within the framework of the model used, the value of θ does not change with a change of the angle ϕ. In this case θ_4 and θ_3 will be connected by elementary geometric relations.

Figure 4a shows the value of W_m computed in this way, as a function of the angle of rotation of the primary beam around the ⟨001⟩ axis of a body-centered cubic lattice for the case U = 260 eV, $\theta_4 = 30°$, $\psi = 126°$. In the calculations we took into account collisions of the ion with four, three, and two lattice atoms (respectively (100) planes, (110) planes, and horizontal lines). The curves of Fig. 4 determine the maximum energies of the scattered ions. It is evident from these curves that W_m has a peak in the region of small positive values of ϕ and changes rather sharply with change of ϕ. From comparison of these curves with the experimental data (Fig. 2) it follows that, in spite of the simplifications made in the calculation and the relatively low resolving power of the apparatus used, satisfactory agreement exists between them.

Figure 4b shows the dependence of $W_m(\phi)$ ($\theta_4 = 30°$) calculated for the same angles of scattering of the primary ions by the (110) plane as those studied experimentally (Fig. 1). In this case also we note a satisfactory agreement of the experimental and theoretical curves. In particular, our attention is drawn to the fact that in both cases with a decrease of ψ the relative "amplitude" of the periodic variations of W_m with ϕ decreases. These facts are an additional argument in favor of the previously assumed model[4,10] of the mechanism of unpaired interaction of a slow ion with the surface of a solid.

A factor which to a considerable degree determines the possibility of such an unpaired collision is the "effective diameter" d_{eff} of the bombarding ion. On the other hand, the possibility of penetration of the primary ion inside the target depends on this same factor,[11] and also the probability of its successive collisions with individual lattice atoms.[12] Therefore, in the study of the role of

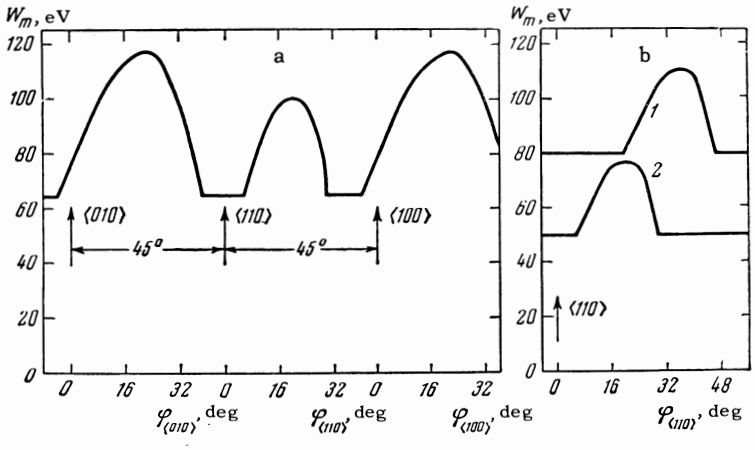

FIG. 4. Theoretical dependence of the maximum energy of the scattered ions on the angle ϕ between the primary ion direction and the normals to the different scattering planes: a – U = 260 eV, $\psi = 126°$, target is rotated around the <001> axis; b – U = 200 eV, $\psi = 90°$ (1) and 126° (2), target is rotated around the $<\bar{1}11>$ axis.

the two collision mechanisms in the ion scattering process, it is of interest to compare the dependences $W_m(\chi)$ obtained in bombardment of the same target by ions having different values of d_{eff}. With this aim we have studied $W_m(\chi)$ for the ions Cs^+, Rb^+, and K^+, scattered from a single crystal of molybdenum.

We have shown previously[10] that to characterize the scattering it is convenient to use the concept of effective mass of the lattice atom M_{eff}, which can be uniquely calculated from the experimentally determined values of W_m (see formula (2)). Figure 5a shows the values of $M_{eff}(\chi)/M$ determined in this way for the case of a molybdenum target rotated around the $\langle 10\bar{1}\rangle$ axis. For Cs^+, two clearly expressed peaks are observed for scattering of ions from the (101) and (010) "planes" and also a small peak possibly due to reflection of the ion from the four lattice atoms (three located far

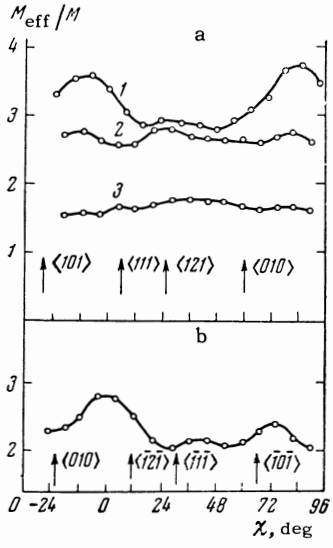

FIG. 5. The ratio M_{eff}/M as a function of the angle of incidence of the primary ions on the surface of molybdenum (a) and tungsten (b) targets rotated around the $<10\bar{1}>$ axis ($\psi = 126°$, U = 260 eV, $\Delta \leq 5°$). Ions: 1 – Cs^+, 2 – Rb^+, 3 – K^+.

from each other in the (111) plane and the fourth lying below them). On changing to Rb^+ ions, the value of d_{eff} decreases. Correspondingly, a considerable reduction in the height of the $\langle 101\rangle$ and $\langle 010\rangle$ peaks occurs, and indications of the occurrence of a $\langle 121\rangle$ peak appear. Finally, for K^+ ions, which have a still smaller d_{eff}, the height of the $\langle 121\rangle$ peak outgrows the heights of all the others, and the process associated with scattering of ions in this plane becomes the process determining the average value of W_m. From considerations of crystal geometry it is clear that we are encountering here a scattering mechanism of a different type from that discussed above. Since the $\langle \bar{1}1\bar{1}\rangle$ direction, which lies in the (121) plane along which the scattering occurs in this case, has a maximum packing density, it is natural to assume that the occurrence of the peak described is the result of the successive scattering of ions by individual lattice atoms.[8]

Comparison of the heights of the series of other peaks on the basis of crystal geometry considerations shows that their occurrence cannot be due to the process of successive binary collisions. Therefore the contribution of this process to the value of $\overline{W}_m$ in the case of Cs^+ and Rb^+ is small.

Figure 5b shows a plot of $W_m(\chi)$ for Cs^+ ions scattered by a tungsten target rotated around its $\langle 10\bar{1}\rangle$ axis. From comparison of Fig. 5b with curve 1 of Fig. 5a, it follows that in spite of the difference in the shape of the scattering potential of the target atoms in the two cases (tungsten and molybdenum), the relative height of the peaks and consequently also the contribution of unpaired collisions remain approximately the same in this case. Possible causes of the difference in absolute values of M_{eff}/M for targets with different ratios of M to m have been discussed by us previously.[9]

From the results reported above we can conclude that in the energy region studied, both binary

and unpaired collisions play a role, the relative contributions of the collisions of the two types depending in great measure on the effective diameter of the scattered ion. The size of the lattice atoms plays a smaller role in this process.

On the basis of the experimental data obtained and the theoretical scheme presented, we have made an estimate of the value of the scattering potential for interaction of alkali-metal ions with atoms of tungsten and molybdenum. The potential turns out to be closer to the Thomas-Fermi-Firsov potential[13] than to the Thomas-Fermi-Dirac potential.[14] More accurate and detailed information on the actual form of this potential will require very tedious calculations with electronic computers[15] and is outside the scope of the present work.

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