Scattering of molecular ions and their atomic fragments by a metal surface

S. A. Evstigneev, S. N. Zvonkov, and V. M. Chicherov

I. V. Kurchatov Atomic Energy Institute
(Submitted 18 November 1981)
Zh. Eksp. Teor. Fiz. **82**, 1096–1103 (April 1982)

Scattering of molecular and atomic ions with energy $(1-2)\times10^4$ eV by a metal surface is investigated. The energy spectra of the scattered ions obtained by bombardment with various ions are measured and compared. It is established that the atoms that make up a molecular ion are scattered differently than the corresponding atomic ions impinging one by one on the metal surface. The differences are attributed to scattering of one of the atoms contained in the molecular ion by another atom of the same ion.

PACS numbers: 79.20.Nc

INTRODUCTION

In experiments on fast-ion scattering by metals the targets are usually bombarded by atomic ions. Molecular ions are extremely rarely used in these experiments. In essence, all that is known of the scattering of fast molecular ions is that they are dissociated by impact against a surface, and the dissociation probability is very high, so that the scattered beam contains a fraction of 1% of the molecular ions and the energy spectra of the scattered ions have sharp peaks that correspond to the scattering of atomic fragments of the molecular ion by the target atoms (see Ref. 1).

The formal picture gives grounds for assuming that the scattering of a fast molecular ion reduces simply to scattering, by the target atoms, of the constituents of this ion, and the latter are individually scattered independently of one another, i.e., as if the corresponding atomic ions were to arrive at this surface one by one. The extent to which this representation is true, however, is not clear. This can be established only by comparing the experimental data obtained by bombarding a surface with molecular ions directly with the results of experiments in which the same target is bombarded by atomic fragments of the considered molecular ions. We do not know of any such studies, with the exception of Ref. 2, where some data are given for hydrogen ions.

We describe below the results of a comparison of the energy distributions of a scattered stream of particles obtained by bombarding targets with molecular ions and their atomic fragments. We used in the experiments various ions and targets of various metals. The revealed regularities have, in our opinion, a sufficiently common character.

EXPERIMENT

The experimental setup is shown in Fig. 1. The apparatus consisted of three main parts: a) device for shaping the beam of bombarding ions, b) a scattering chamber with a set of targets, c) an energy analyzer for the scattered ions.

The bombarding-ion beams both the main one used for measurement and the auxiliary one used to clean the target surface, were produced by ion sources of the duoplasmatron type. The separations of the ions of the required species and the shaping of the main beam were effected with a magnetic mass separator with average-trajectory radius 25 cm and rotation angle 90°. The energy of the bombarding ions was varied in the interval from 5 to 30 keV.

The scattering chamber, 400 mm in diameter and 250 mm high, was made of stainless steel. It was evacuated with a titanium getter pump and a diffusion pump. The pressure of the residual gases in the scattering chamber was usually $(1-2)\times10^{-8}$ Torr. The target was placed at the center of the chamber. Its surface was cleaned before each measurement by bombardment with argon ions from the auxiliary ion source.

The radius of the equilibrium trajectory of the electrostatic analyzer was 450 mm, and the deflection angle was approximately 150°. The height of the analyzer slits was 10 mm, and their widths could be varied from 1 to 20 mm. The energy line width at the analyzer output, when the ion beam from the mass separator was incident on its input slit, was approximately 0.3% (see Fig. 3). The electrostatic analyzer was connected to one of the vacuum nipples located on the lateral cylindrical surface of the scattering chamber. This made it possible to select a particular scattering angle.

The ions were registered with an open electron multiplier, while the spectra were registered with an automatic x-y recorder.

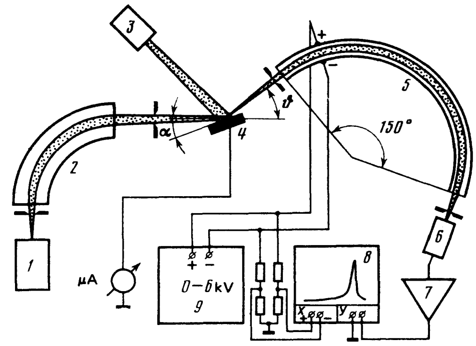

FIG. 1. Experimental setup: 1) source of bombarding ions, 2) separating magnet, 3) auxiliary ion source, 4) target, 5) electrostatic energy analyzer, 6) detector, 7) preamplifier, 8) automatic x-y recorder, 9) analyzer power supply.

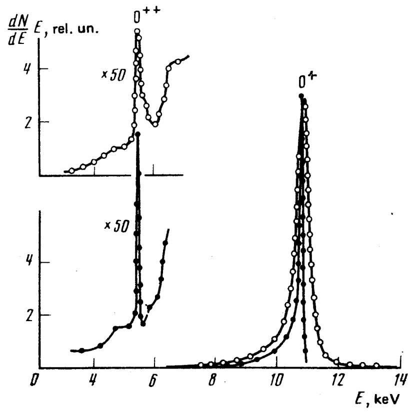

FIG. 2. Energy spectra of scattered ions in bombardment of a gold target by the following ions: O^+ with energy 11.43 keV (•), O_2^+ ions with energy 22.86 keV (○); $\vartheta=40°$, $\alpha=20°$. The peaks of the singly scattered O^+ ions were made of equal height.

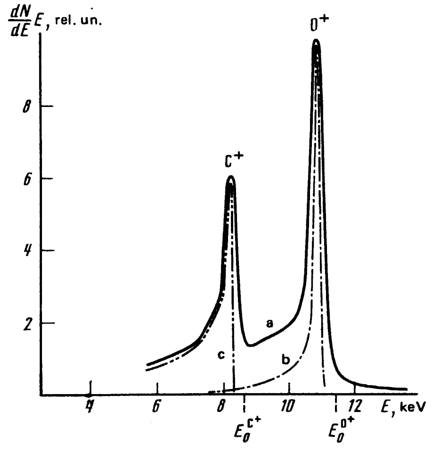

FIG. 4. Energy spectra of the scattered ions in bombardment of a gold target by the following ions: a) CO^+ with energy 10 keV, b) O^+ with energy 11.4 keV, c) C^+ with energy 8.6 keV. The peaks of like ions are equalized in height; $\alpha=20°$, $\vartheta=40°$.

The targets were bombarded by beams of singly charged molecular ions N_2^+, O_2^+, CO^+, CO_2^+ and by beams of atomic ions N^+, C^+, O^+, which were fragments of these molecular ions. The energy of the bombarding molecular ions was in most cases 20 keV, while that of the atomic ions was chosen every time to equal the kinetic energy possessed by the given ion when it bombarded the target and was part of the molecular ion of the corresponding species. For example, for comparison with the spectra obtained in bombardment by CO_2^+ ions, we used C^+ and O^+ ions with respective energies 6 and 8 keV.

Typical energy spectra are shown in Figs. 2–7. When the bombardment was by atomic ions, the spectra obtained were quite ordinary: they contained peaks of beam ions scattering without stripping, peaks of beam ions scattered with stripping, and peaks of ionized atoms of the target–recoil atoms produced in pair collisions with the incident ion.

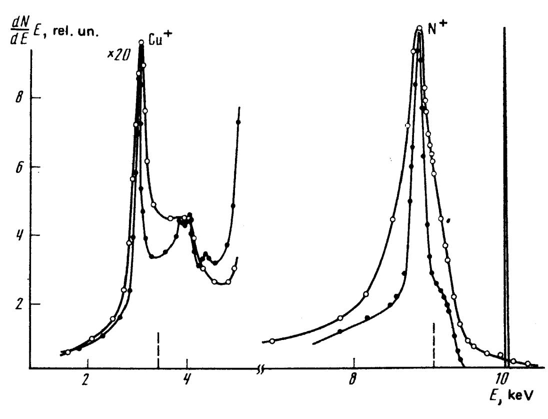

FIG. 3. Energy spectra of the scattered ions in bombardment of a copper target by the following ions: N^+ with energy 10 keV (•), N_2^+ with energy 20 keV (○); $\alpha=20°$, $\vartheta=40°$. The peaks of the singly scattered N^+ ions are made equal in height. On the right side is the energy spectrum of the bombarding ions. The dashed lines show the calculated positions of the peaks without inelastic-collision angles.

In the case of bombardment by molecular ions, the spectrum contained peaks corresponding to scattering of their atomic fragments by metal atoms and to scattering both without and with stripping, as well as peaks of recoil atoms produced in pair collisions with these fragments. The number of molecular ions unaffected by scattering was negligibly small (at scattering angles larger than 15–20°).

If the target is bombarded by homonuclear molecular ions, say O_2^+ (Fig. 2) or N_2^+ (Fig. 3), the spectra contain a sharp peak corresponding formally to scattering of O^+ ions of energy half the initial energy of the molecular ion by the target. In experiments with non-homonuclear ions, e.g., CO^+ or CO_2^+ (Figs. 4 and 5), the spectra contain peaks corresponding to scattering of each atomic fragment, i.e., of the O^+ and C^+ ions, by the target atoms. The crests of all the peaks observed in scattering of molecular ions are close to the positions of the crests of the peaks observed in the energy spectra in the scattering of the corresponding atomic ions, although full coincidence was not observed in all cases.

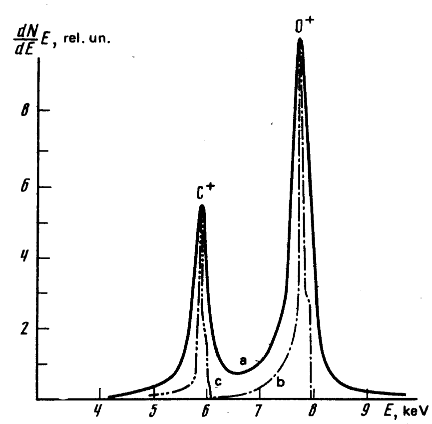

FIG. 5. Energy spectra of scattered ions in bombardment of a gold target by the following ions: a) CO_2^+ with energy 22 keV, b) O^+ with energy 8 keV; c) C^+ with energy 6 keV. The peaks of like ions are equalized in height; $\alpha=20°$, $\vartheta=40°$.

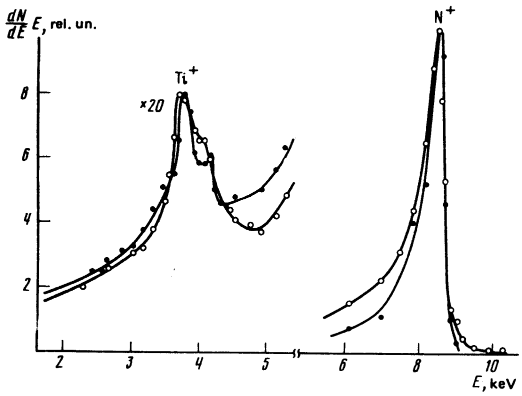

FIG. 6. Energy spectra of scattered ions in bombardment of a titanium target by nitrogen ions: N_2^+ with energy 20 keV (○) and N^+ with energy 10 keV (●). The peaks are equalized in height; $\alpha = 20°$, $\vartheta = 40°$.

The shapes of all the peaks in the spectrum depend substantially on which ions, molecular or atomic, bombard the target. In the spectra obtained by bombardment with atomic ions, the right-hand high-energy slope of the peak of the "singly" scattered ions of the beam approaches the abscissa axis steeply, almost perpendicularly, so that its intensity decreases rapidly to a vanishingly low value. In experiments with molecular ions, the right-hand slope of the single scattering peak approaches the abscissa axis slowly and terminates in a long tail. This tail extends towards higher energies so far that a noticeable intensity is observed even at energies greatly exceeding the initial kinetic energy possessed by the corresponding ion when it bombarded the target while still part of the molecular ion.

The peaks of the recoil atoms and the peaks corresponding to scattering of the beam ions with stripping differ greatly in these two cases. For example, the peak of O^{++} ions (Fig. 2) has a clearly pronounced ledge on the right in the case of bombardment by molecular ions, whereas in the case of atomic bombardment this ledge vanishes and the peak itself is much narrower. We note that in this spectrum there are no

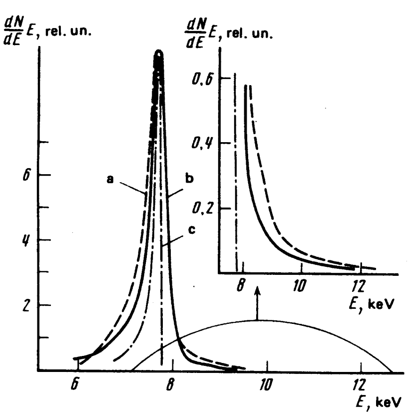

FIG. 7. Energy spectra of scattered ions in bombardment of a gold target by nitrogen ions: a) N_2^+ with energy 24 keV, b) N_2^+ with energy 16 keV, and c) N^+ with energy 8 keV. The peaks are equalized in height; $\alpha = 20°$, $\vartheta = 40°$.

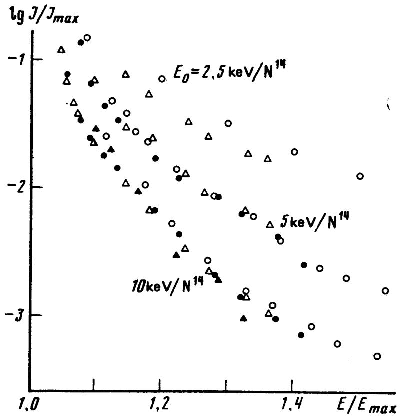

FIG. 8. High-energy sections of spectra in bombardment of targets of different metals by N_2^+ ions (○—Cu, ●—Rh, △—Au, ▲—Ti). The "tails" of the energy spectra are plotted in such a way that their heights are equal at the main peaks. E_{max} is the energy corresponding to the position of the main peak.

ledges whatever on the right-hand slope of the peaks of the ions scattered without stripping. The O^{++} peak in the spectra where the O^+ peaks are equalized in height is almost twice as high in the case of atomic bombardment, but the areas under these peaks are approximately equal.

The peak widths corresponding to scattering of atomic and molecular ions differ greatly in the case of heavy targets, such as gold or rhodium; these differences vanish on going to targets with small Z (cf. Fig. 7 with 3 and 6). Thus, e.g., the spectra obtained by bombarding a graphite target with molecular and atomic nitrogen ions hardly differed from each other.

In contrast to the foregoing, the target material has practically no effect on the high-energy part of the spectrum, i.e., on the tails adjacent on the right side to the single-scattering peaks in the case of bombardment by molecular ions. To emphasize this circumstance, Fig. 8 shows in a logarithmic scale the high-energy sections of the spectra obtained by bombarding targets of different metals.

As seen from the same figure, the lengths of the energy-distribution tails are strongly influenced by the energy of the bombarding ions: the experimental points obtained for different targets at three values of the primary ion energy are distinctly bunched near three curves corresponding to these energy, regardless of the bombarded target. On the contrary, the ratio of the widths of the peaks of the scattered ions is not noticeably influenced by a change in the primary-ion energy.

DISCUSSION

Thus, a detailed comparison of the energy spectra obtained by bombarding a number of targets, in one case by molecular and in the other by atomic ions, demonstrates the pronounced difference between these spectra. The difference are connected with the singularities of the scattering of the molecular ions. In most general outline, these singularities lead to the appearance of scattered particles that have an energy higher as well as lower than that they would possess if the scattering of the atoms constituting the molecular ion at the instant of scattering, were perfectly analogous to the scattering of the corresponding atomic ions arriving at the surface of the metal one by one. As follows from a comparison of the energy spectra, for most scattering particles the absolute value of the observed energy difference is not particularly large. This is evidence that the principal part of the interaction between atoms combined into a molecular ion and metal surface is scattering of each of these atoms by the target atoms, just as in the case of bombardment by atomic ions. In contrast to atomic ions, however, atoms in molecular ions have some additional possibility of momentum exchange. This possibility might be provided by scattering of one of the atoms in the molecular ion by its other atom, which arrives at the target surface first and slows down earlier. Let us refine this, at first glance primitive, mechanical model of the evolving process. Scattering of one of the atoms contained in a molecular ion (we call this the second atom) by the other, first atom can either follow the scattering of this second atom by the metal atom, precede it, or else be intermediate between the two successive scatterings of the second atom by the metal atoms. It is important to note that this additional scattering of one atom of the molecular ion by its other atom can, in principle, make possible those double collisions of the incident atom with the metal atoms, which were impossible in the absence of this additional scattering (i.e., in case of bombardment by atomic ions).

Elementary calculations of the energy loss in elastic paired collisions can show that this additional scattering of constituent atoms of a molecular ion by each other can in principle lead to either a decrease or an increase (if paired double collisions with the metal atoms begin to be realized) of the scattered-ion energy compared with the energy possessed by the ion scattered only by one metal atom. It turns out that these energy changes are such that in principle they can explain the observed broadening of the spectra.

The existence of high-energy tails of the spectra cannot be attributed to an increase of the multiplicity of the collisions with the metal atoms. To explain them it must be assumed that in the course of the scattering by the target atoms a considerable momentum transfer takes place from one of the molecular-ion atoms to the other.

The authors are grateful to S. Yu. Luk'yanov for support and interest, and to O. B. Firsov and Yu. V. Martynenko for helpful discussions.

[1]E. S. Mashkova and V. A. Molchanov, Rasseyanie ionov srednikh energiĭ poverkhnostyami tverdykh tel (Scattering of Medium-Energy Ions by Solid Surfaces), Atomizdat, 1980, p. 91.

[2]W. Heiland, U. Beitat, and E. Taglauer, Phys. Rev. **B19**, 1667 (1979).

Translated by J. G. Adashko