Fast electrons from slow atomic collisions

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We report observations of extremely energetic electrons produced in collisions between keV He^+ and Ar^+ and different solid surfaces, with energies up to approximately 40% of the center-of-mass energy. This emission can result from the decay of many-electron excitations produced in nearly head-on collisions.

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Ionization is one of the most prominent but least understood processes occurring during collisions of atomic particles at velocities smaller than the orbital electron velocities of the colliding partners. Much of the substantial experimental data that exist for total cross sections and low-energy electron spectra are not described by current theories. At these low velocities, the collision is usually described in terms of molecular states of the quasimolecule which is formed transiently during the atomic encounter. Electronic excitations are thought to occur through perturbations of quasimolecular states by the atomic motion. Often, electronic excitations are described by one-electron transitions between molecular orbitals in the quasimolecule. Individual mechanisms which have been proposed to account for ionization at low velocities include direct coupling to the continuum and inner-shell excitation followed by Auger decay during or after the collision. A few early theories [1] propose multielectron excitation mechanisms but their development ceased when the successful one-electron promotion models appeared. Although one-electron theories are successful in explaining inner-shell excitations they fail to explain multiple ionizations. A question which is still unsolved is the mechanism for production of energetic continuum electrons. Like others [2–5], we have long been puzzled by these continuum electron spectra ("continuum tails") which exist at energies higher than Auger-electron spectra [6,7]. The origin of these tails is a matter of current debate [8]; the continuum has been interpreted in some cases to result from the decay of quasimolecular autoionizing states during the collision, or from direct coupling of a bound state to the continuum.

Previous observations on electron continuum tails have been restricted to relatively small electron energies, usually less than 1% of the projectile energy. To see how far these tails extend, one needs more sensitivity; this can be achieved with solid targets due to their large density of target atoms. We do not expect any essential effect in the primary ionization that is specific to the solid state but rather that the electrons originate from single collisions. This is because (i) very inelastic collisions occur at small internuclear distances, where the effect of the neighbors is relatively unimportant; (ii) the electrons emitted with high energies can only travel a few nm inside the solid; it is expected that the energy degradation of the projectile in a region of this thickness is not very important, and (iii) electron ejection occurs fast, before any appreciable relaxation of the valence electrons in the solid can take place. Compared with gas-phase collisions, the solid provides additional energy-loss mechanisms which produce energy-dependent attenuation [9] of the flux of fast electrons.

Our measurements for keV ions in solids show that the continuum-electron energy distributions extend up to a surprisingly large fraction of the center-of-mass energy $E_{c.m.}$. This indicates that the high electron energies result from nearly head-on collisions between the projectile and a target atom. In some cases, we have seen indications of a maximum energy of the ejected electrons.

The measurements were preformed in the energy range 0.5–6 keV, with singly charged He and Ar ions on clean targets of Be, Mg, Al, Si, Cu, Ce, and Au at three different laboratories (Bariloche, Rutgers, and Cosenza), which are equipped with different electrostatic electron energy analyzers operating under ultrahigh vacuum. The system in Bariloche [10] has a CLAM hemispherical analyzer, made by Vacuum Generators (VG). At Rutgers University, the setup [11] uses a VSW HA50 hemispherical analyzer which has a hole in the outer hemisphere to prevent scattered neutrals and photons from ejecting electrons from this plate. Both instruments could be operated by scanning the voltage between the hemispheres or by fixing this voltage and scanning a potential to retard the electrons before entering the analyzer. These instruments also have a lens which images the beam spot (electron source) onto the entrance slit of the analyzer. In Cosenza, we used a Varian cylindrical-mirror energy analyzer. In some cases the count time per channel was set proportional to ion energy to obtain better statistics at high energies. The spectra were then corrected for this; in all cases the spectra were corrected for the energy-dependent analyzer transmission. The data shown here were taken with the VG instrument aimed 15° from the normal to the surface and at 45° incidence of the ion beam. Very similar data were obtained with the other two instruments for Mg, Al, and Si targets, showing that the results are not very sensitive to the angle of ion incidence or the angle of emission of the electrons.

The log-scale plot makes the high-energy tail conspicuous, as shown in Fig. 1 for Ar on Mg, with the incident energy as a parameter. We find electrons with extremely high energies, up to $\sim 40\%$ of $E_{\text{c.m.}}$. The tails decay exponentially and a high-energy cutoff is apparent in the case of Au (Fig. 2). The large fraction of $E_{\text{c.m.}}$ which goes into electronic excitation must be accompanied by a large change in momentum, which can only occur in nearly head-on collisions between the projectile and a target atom. Figure 2 shows that fast electrons also occur for light-ion impact, although with a smaller probability. The high-energy cutoff occurs at somewhat smaller energies than for Ar ions at the same impact energy. In both cases, the energy cutoff grows approximately linear with impact energy.

Figure 3 shows data for Ar impact on different targets. One can notice a strong dependence of the high-energy tail with the target atomic number, with the tail being more pronounced for the heavier targets. The data for Ce shows that the behavior is not monotonic with the atomic number of the target.

We have been concerned about possible experimental artifacts that could account for the observations. This was the motivation for doing experiments in different laboratories with different types of instruments operating under different geometrical arrangements. We found that the shape and extent of the high-energy tails were not affected by operating the hemispherical analyzers with retardation of the electrons before analysis. The spectra were not affected by biasing the channeltron detector to prevent collection of low-energy electrons which could conceivably originate from particle scattering in the plates. Furthermore, we tested both hemispherical analyzers by measuring the energy distributions of electrons under primary electron impact. In this case, the feature with the highest energy in the spectra is the peak of elastically scattered electrons. We observed that this peak decays exponentially on the high-energy side, disappearing into the background a few eV away from the peak energy. This behavior has been observed for electrons of energies in the range 50 eV–3 keV and shows that the signals appearing at high analyzer pass energies do not originate from low-energy electrons hitting internal surfaces of the analyzer. It should be pointed out that the input lens in these analyzers acts as an energy prefilter reducing the background due to unwanted electrons [12].

Another possibility is that electrons are ejected inside

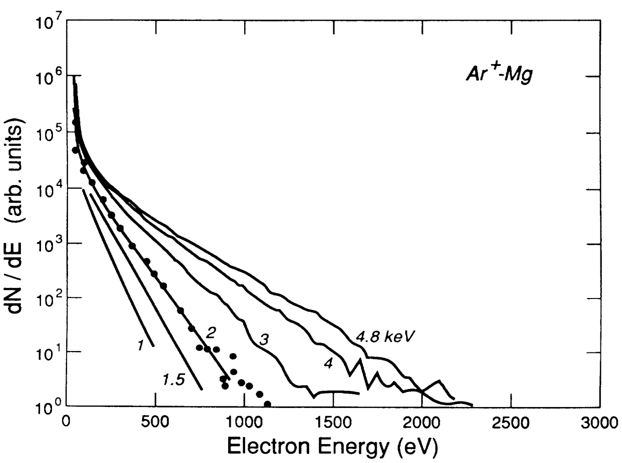

FIG. 1. Electron energy spectra of Mg under bombardment with Ar^+ ions at different ion energies. Data points shown are for 2-keV impact energy.

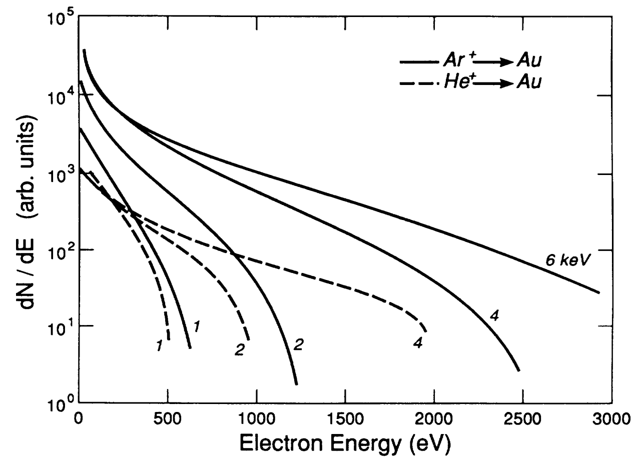

FIG. 2. Electron energy spectra of Au under He^+ and Ar^+, as a function of projectile energy.

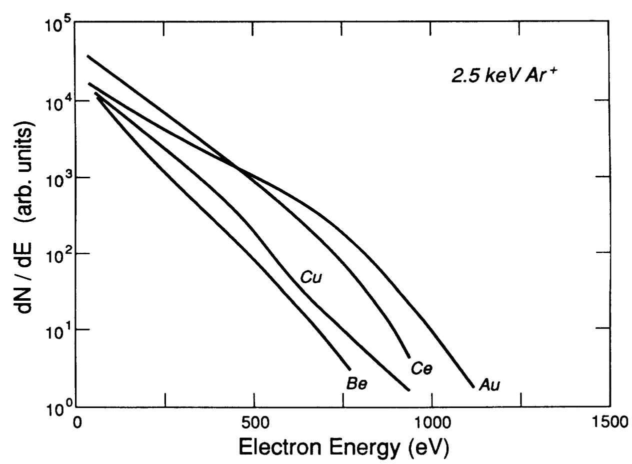

FIG. 3. Energy spectra of several targets under 2.5-keV Ar^+-ion bombardment.

the analyzer by backscattered heavy particles. In the VSW spectrometer, energetic neutrals cannot hit the outer hemisphere, unlike the case of the VG instruments. Both analyzers gave essentially identical spectra. This finding, and the fact that the shape of the tails does not depend on the use of preretardation of the electrons before analysis, show that the emission of stray electrons by backscattered neutrals is not important.

The tails cannot be produced by energetic negative ions. Negative backscattered projectiles can only occur for He, since Ar does not form long-lived negative ions. Furthermore, the spectra do not resemble the energy spectra of backscattered ions. Neither can negative *target* ions account for the observations, since the measured energies are much larger than those which can be transferred in a violent collision, particularly in a He-Au collision.

We thus conclude that the signals observed are due to high-energy electrons. The origin of these electrons has not as yet been clarified. Any model for the process must take into account or explain the following crucial facts.

(1) The transfer of a substantial fraction of the center-of-mass energy to a single electron. Conservation of energy and momentum dictates that this excitation occurs in a head-on or nearly head-on collision.

(2) The existence of a high-energy cutoff in the continuum tail, and its energy dependence.

(3) The occurrence of high-energy electrons both for light and heavy projectiles.

(4) The dependence of the high-energy tail with target atomic number.

The conversion of a large fraction of the center-of-mass energy to electronic energy also occurs near the ionization threshold. Amme and co-workers [13] have found structure in the ionization cross section of Ar and Kr near threshold which suggests that complex excitations become important as the impact energy increases. We then advance a qualitative model where electron emission results from the decay of a many-electron excited quasimolecular state formed during nonadiabatic compression of the electron clouds in the collision. This is conceptually similar to the single electron decay of a plasmon excitation in solids [14]. On approach, the electron clouds of the projectile and target are compressed and multiple electron promotion occurs due to Pauli excitation [15], through nonadiabatic crossing of molecular orbitals. At the turning point, the velocity of relative motion drops near zero, and the system has time to relax by many electron transitions. Statistically [3], this rearrangement can also occur by ejecting a single electron. Although the model is qualitative, it rationalizes the main features observed. For instance, the degree of inelasticity increases with particle energy, since deeper shells become available for excitation and since the distance of closest approach diminishes, forcing electrons to occupy a smaller volume. The tails are more prominent the heavier the projectile and target atom, which is consistent with a higher number of filled orbitals which are promoted. The many-electron nature of the excitation could also explain why simple Auger transitions are not observed from deep inner shells which could conceivably be excited in such deep inelastic collisions.

The production of highly energetic electrons is likely to be related to the yet unexplained threshold behavior of ion-induced electron emission from solids [16]. Another manifestation of these highly inelastic collisions should be the appearance of long tails in the energy-loss distributions of ions after single atomic collisions or upon multiple collisions with solid matter.

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