Ionization of K(*nd*) Rydberg-state atoms at a surface

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The ion production that results when K(*nd*) Rydberg atoms are incident at grazing incidence on a metal surface is investigated. The data show that such ion production forms the basis of a simple technique with which to detect Rydberg atoms, even at relatively low values of n, and suggest that Rydberg atoms may provide a novel means to investigate local electric fields present near a surface.

Atoms in high-lying Rydberg states incident on a metal surface can be ionized through tunneling of the excited electron into a vacant level in the metal.[1-4] Theoretical studies by Chaplik[2] indicate that the tunneling rate increases very rapidly with decreasing atom-surface separation and suggest that ionization will occur over a narrow range of atom-surface separations at $\sim(4-5)n^2a_0$, where n is the principal quantum number of the incident Rydberg atom and a_0 is the Bohr radius. An ionization distance of $\sim 4.5n^2a_0$ was inferred by Fabre *et al.*[3] from measurements of the transmission of Na(*nd*) Rydberg atoms through an array of micrometer-size slits by assuming that the observed decrease in transmission with increasing n resulted from surface ionization at the edges of the slits. Recent work by Kocher and Taylor[4] using a fine-mesh grid and lithium Rydberg atoms, however, indicates that the transmission might also be affected by localized electric fields produced by absorbed surface layers in the vicinity of the holes. In neither transmission experiment was ion production investigated. In the present work we have observed directly the ion formation that results when K(*nd*) Rydberg atoms are incident at a near-grazing angle on a metal surface. The data demonstrate not only that such ion formation can form the basis of a simple technique with which to detect Rydberg atoms but also point to the existence of localized electric fields near the surface, suggesting the use of Rydberg atoms as a surface probe.

The present apparatus is shown schematically in Fig. 1. Potassium atoms contained in a tightly collimated thermal-energy beam are excited, in zero electric field in an electrostatically shielded region, to a selected *nd* state by two-photon excitation using the focused output of a Coherent Radiation Inc. Model 699-21 ring dye laser. The Rydberg atoms exit the excitation region and are incident at an angle of 2° on the surface of a plane gold mirror. An electric field is established perpendicular to the surface by biassing a fine-mesh grid located 2 mm above the surface. This field accelerates ions from the surface through a further fine-mesh grid and into a second acceleration region. Both fine-mesh grids are operated at the same potential; the second grid is used to minimize any possible effects due to field penetration. In the second acceleration region the ions are accelerated by a uniform electric field to an energy of 1 keV prior to detection by a position-sensitive detector (PSD).[5] Figure 1 includes a typical distribution of ion arrival positions across the PSD. Significant ion production is only observed from regions of the surface impacted by Rydberg atoms.

As apparent from Fig. 1, the gold mirror is subject to potassium deposition. Attempts to minimize the buildup of potassium on the surface by operating with it heated to ~ 400 °C proved unsuccessful due to a large background of thermally desorbed ions. (The surface was, however, routinely baked to 450 °C between data runs.) The data presented here were recorded with the surface maintained at $\sim 80-100$ °C. This temperature is above the melting point of potassium, thereby preventing the growth of microcrystallites on the surface, but is insufficient to prevent potassium deposition. Further, potassium on the surface may react with the background gas (pressure $\sim 5\times 10^{-8}$ Torr). Nevertheless, although the surface conditions are not well defined, the features and characteristics of the data obtained were quite reproducible.

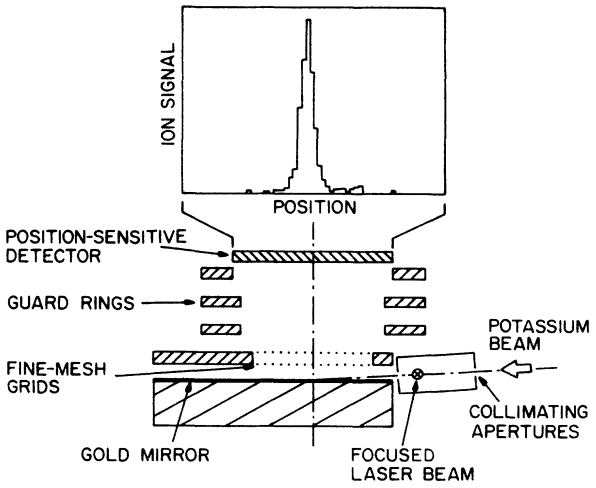

FIG. 1. Schematic diagram of the apparatus. Inset shows the distribution of ion arrival positions across the position-sensitive detector for incident K(20*d*) atoms.

The dependence of the measured ion signals on the electric field applied perpendicular to the surface is illustrated in Fig. 2 for incident atoms in several *nd* states. The observed ion signals have a sharp onset near zero applied field and then increase with increasing field. The location of the onsets, and their lack of a dependence on n, was quite unexpected. Experiments have shown that alkali-metal ions striking a low-work-function surface such as produced by alkali-metal-atom deposition are efficiently neutralized.[6,7] An ion formed near a conducting surface experiences an attractive electric field due to its image charge. At small ion-surface separations, this field can be quite large. For example, the ion formed by ionization of a 20*d* atom at an atom-surface separation of $\sim 5n^2a_0$, i.e., ~ 106 nm, experiences an initial electric field of ~ 320 V cm^{-1}. This suggests that, in the absence of other fields, the ion will be rapidly accelerated toward the surface. Thus it was anticipated that, in order to observe an ion signal, a sizeable external electric field would have to be applied perpendicular to the surface to prevent ions from striking the surface and being lost. Calculations of ion trajectories under the joint influence of external and image charge fields showed that, because the incident Rydberg atoms have only small components of velocity normal to the surface, the probability that ions strike the surface decreases from unity to zero over a narrow range of critical external fields, the exact values of which depend on the distance from the surface at which the ions were initially formed (thus providing a means to measure the ionization distance). Figure 2 includes a series of horizontal bars that indicate, for each value of n, the range of critical fields over which the onset in ion signal was anticipated, assuming that ionization occurs at atom-surface separations in the range $(4-5)n^2a_0$. In each case these fields are significantly lower than those necessary for direct field ionization.[8]

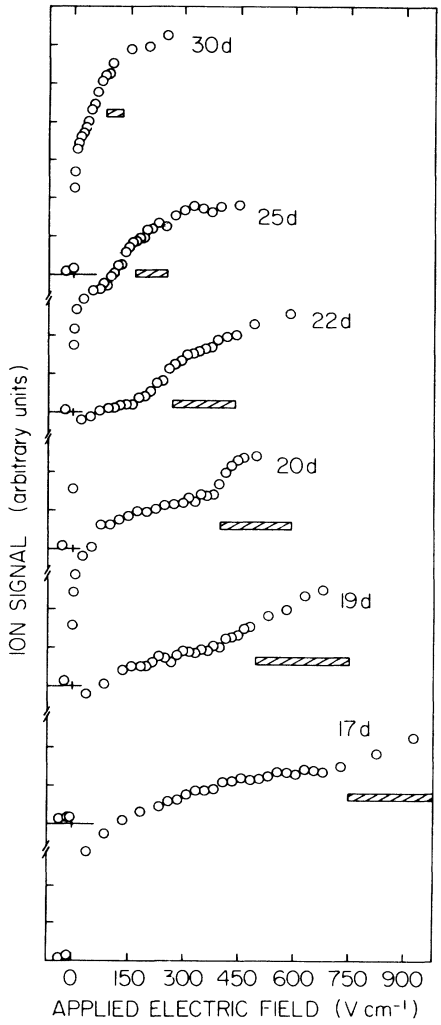

FIG. 2. Dependence of the measured ion signal on electric field applied perpendicular to the surface. Horizontal bars indicate, for each value of n, the range of fields over which the onset in the ion signal was expected (see text).

As is apparent from Fig. 2, ion signals are observed at applied fields that are very much smaller than initially expected. This cannot be explained by invoking ionization at very large atom-surface separations because the ionization distances required are much too great to be compatible with earlier studies of Rydberg-atom transmission through slits or grids.[3,4] It is also unlikely that the observed low-field onsets results from interactions involving atoms in states of very high n produced by blackbody-radiation-induced photoexcitation from the parent *nd* state. The anticipated photoexcitation rates are too small to cause significant population transfer during the flight time from the excitation region to the surface, typically ~ 35 μsec. The ion signal is also not due to blackbody-radiation-induced photoionization. This process would result in an ion signal that decreases exponentially between the excitation region and the point where the Rydberg-atom beam impacts the surface. As evident from Fig. 1, the ions observed in the present experiment originate from a small area on the mirror surface that coincides with the expected impact positions of the excited atoms. Taken together, these considerations indicate that the locations of the observed onsets are governed by surface properties (an idea further reinforced by their n independence). One possible explanation for the small onset fields observed is that localized electric fields exist near the surface that, over a fraction of the surface, are of sufficient magnitude and in the correct direction to counteract the field due to the image charge, thereby permitting some ion escape even at small values of applied field. Such localized fields could result from variations in work function across the surface[9] due to, for example, unequal potassium deposition rates at different points on the surface caused by surface irregularities.

The behavior of the ion signal as a function of applied field above the onset is also consistent with the existence of local electric fields near the surface. It is reasonable to expect that such fields will be quite nonuniform, varying in both magnitude and direction across the surface. In this event, the effect of an externally applied field is to increase the fraction of the surface over which the *net* electric field is sufficient to allow ion escape, suggesting a steady but sizeable increase in observed ion signal with applied field. The presence of an applied field, however, also changes the lifetimes of the incident atoms, thereby

changing the fraction of the laser-excited atoms that survive passage to the surface and that are therefore able to contribute to the ion signal. Subsidiary experiments in a second apparatus showed that the effective lifetimes of nd states are reduced by the presence of an electric field. (For example, in the case of $25d$ atoms a field of 100 $V\,cm^{-1}$ decreases the effective lifetime from ~ 21 to ~ 15 μsec.) Application of the external field thus decreases the number of excited atoms that strike the surface, partially offsetting the effect of higher ion escape probabilities. It is interesting to note, however, that a significant increase in ion signal is apparent at applied field strengths that correspond to ionization at atom-surface separations of $\sim(4-5)n^2a_0$.

The data point to the existence of sizeable electric fields near the present surface that play an important role in ion escape from the surface. To further explore and understand the ion production and escape mechanisms, a UHV experiment using rare-gas Rydberg atoms is being designed that will provide more carefully controlled surface conditions. Nonetheless, the present work shows that ionization at a surface provides a simple means to detect Rydberg atoms, even at relatively low values of n where detection by field ionization is difficult, and the data suggest that studies with Rydberg atoms may provide a novel means to investigate local electric fields existing near a surface.

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[8]See, for example, T. H. Jeys, G. W. Foltz, K. A. Smith, E. J. Beiting, F. G. Kellert, F. B. Dunning, and R. F. Stebbings, Phys. Rev. Lett. **44**, 390 (1980).

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