Measurement of the Auger Decay after Resonance Excitation of Xe 4*d* and Kr 3*d* Resonance Lines

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The $N_{4,5}O_{2,3}O_{2,3}$ Auger spectra from Xe and the $M_{4,5}N_{2,3}N_{2,3}$ Auger spectra from Kr are investigated for different photon energies around threshold ionization. When exciting at the resonance line ($4d^9 5s^2 5p^6 6p$ for Xe and $3d^9 4s^2 4p^6 5p$ for Kr) we observe the usual Auger multiplet structure to be shifted to higher kinetic energies. Additionally, new lines appear which can be assigned to shake-up processes in the Xe^+ and Kr^+ ions.

Auger spectra usually are obtained for excitation energies (photons or electrons) above threshold of ionization. The $N_{4,5}O_{2,3}O_{2,3}$ Auger spectrum of Xe and the $M_{4,5}N_{2,3}N_{2,3}$ Auger spectrum of Kr have been analyzed.[1-3] Those spectra are rather independent of the excitation energy at photon energies far above threshold. When the excitation energy is lowered to a few eV above threshold, post-collision interaction (PCI) effects due to a nonnegligible coupling of the electron leaving the atom with the rest of the system are observed.[3-5] An even stronger coupling occurs when the atom is excited to a resonance state below threshold. Such a state will decay either radiatively or via an Auger process which can be considered as a special type of autoionization.

We present here the first photoemission measurements of the Auger decay after excitation of Xe to the $4d^9 5s^2 5p^6 (^2D_{5/2})6p$ and to the $4d^9 5s^2 5p^6 (^2D_{3/2})6p$ resonance states at 65 and 67 eV, respectively, and after excitation of Kr to the $3d^9 4s^2 4p^6 (^2D_{5/2})5p$ and $3d^9 4s^2 4p^6 (^2D_{3/2})5p$ states at 91.2 and 92.4 eV, respectively. Such investigations could not be carried out before the availability of a strong tunable light source like synchrotron radiation. They have considerable relevance for a detailed knowledge of the mechanism of deexcitation of certain resonance excitations and also towards a better understanding of correlation effects in excited atoms.

Synchrotron radiation from the storage ring DORIS was monochromatized by the grazing-incidence monochromator FLIPPER.[6] The band pass of the monochromator was set at 0.3 eV. Xenon was used at a pressure of about 1×10^{-4} Torr in a directed gas beam. The photoelectrons were analyzed with a commercial double-pass cylindrical mirror analyzer at a cone with a 2π azimuthal and a 12° polar acceptance. The axis of the cone was perpendicular to the direction of the photon beam and at 45° relative to the electric vector of polarization. These parameters are fairly unimportant for measurements of Auger lines. Especially the β parameter, which is important for the observation of directly excited photoelectrons, has no relevance. The analyzer was used with a band pass of 0.3 eV. The counting rate was typically 30 pulses/sec in the Auger maxima.

In Fig. 1 we show the absorption spectrum of Xe in the region of the onset of the 4*d* transitions. The resonance lines are superimposed on a background of 5*p* and 5*s* electron excitations. Figure

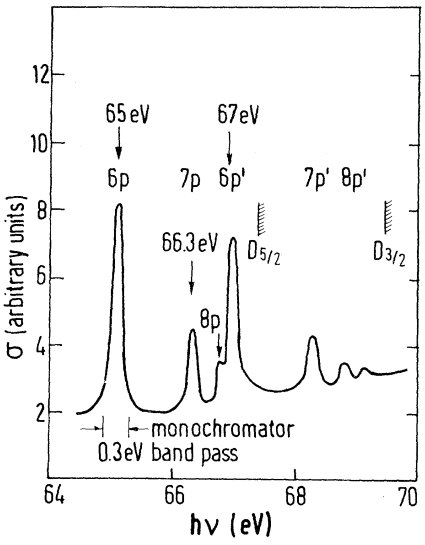

FIG. 1. Cross section vs photon energy for xenon at the onset of the 4*d* electron excitation measured with a resolution of 0.04 eV. The indicated monochromator band pass of 0.3 eV shows the energy width used for the Auger measurements.

1 serves as a reference for illustrating the different excitation processes we have used in the investigations of the Auger decay described as follows. The $N_{4,5}O_{2,3}O_{2,3}$ part of the whole Auger spectrum of Xe taken at 76.7 eV photon energy (an energy well above the ionization threshold for the $4d$ shell) is shown in Fig. 2(a). There are two groups corresponding to transitions from vacancies in the N_4 and N_5 subshells. In view of the resonance excitation, only the peaks originating from a N_5 hole are labeled according to Othani *et al.*[3] Figure 2(b) shows the Auger spectrum after excitation to the $4d^9 5s^2 5p^6 (^2D_{5/2}) 6p$ resonance state at 65 eV. In analogy, Fig. 3 presents the $M_{4,5}N_{2,3}N_{2,3}$ part of the whole Auger spectrum of Kr taken at 117.5 eV photon energy and after excitation to the $3d^9 4s^2 4p^6 (^2D_{5/2}) 5p$ resonance state at 91.2 eV. In addition to the Auger lines the spectra obtained with resonance excitation show the Xe $5s$ and Kr $4s$ lines of the directly emitted photoelectrons.

In the case of resonance excitation there is an overall shift to higher kinetic energies. This energy difference is mainly to be understood as an energy gained from the increase in binding energy of the $6p$ electron for Xe and the $5p$ electron for Kr, due to the formation of a double vacancy in the $5p$ (Xe) and the $4p$ (Kr) subshells. The energies (E_f) of the different final states of the configurations Xe $4d^{10}5s^2 5p^4 6p$ and Kr $3d^{10}4s^2 4p^4 5p$ are obtained from Moore.[7] With the example of the $5s^2 5p^4 (^1S) 6p$ state of Xe^+ we want to explain our considerations. An energy separation of 17.4 eV is obtained between the states $5s^2 5p^5 (^2P_{3/2})$ and $5s^2 5p^4 (^1S) 6p$. Together with the ionization energy of a $5p_{3/2}$ electron in neutral Xe (12.1 eV) we obtain a total energy of 29.5 eV for the $5s^2 5p^4 (^1S) 6p$ final state. The kinetic energies E_{kin} of the emitted electrons are given by the difference between

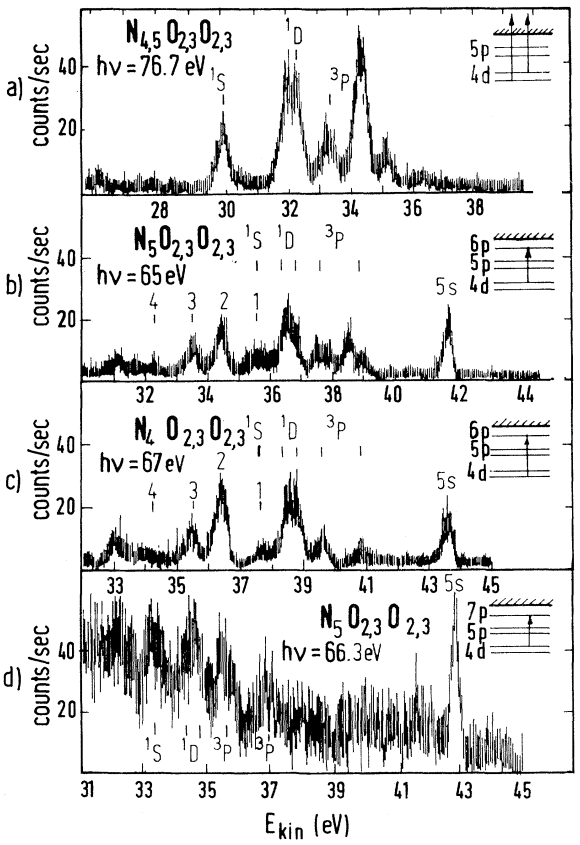

FIG. 2. *NOO* Auger spectra of Xe taken at different photon energies: (a) above threshold of ionization of the $4d$ shell, (b) at the energy for the $6p$ resonance excitation of the $4d_{5/2}$ shell, (c) at the energy for the $6p$ resonance excitation of the $4d_{3/2}$ shell, and (d) at the energy for the $7p$ resonance excitation of the $4d_{5/2}$ shell. The peak labeled $5s$ is due to the direct ejection of electrons from the $5s$ shell.

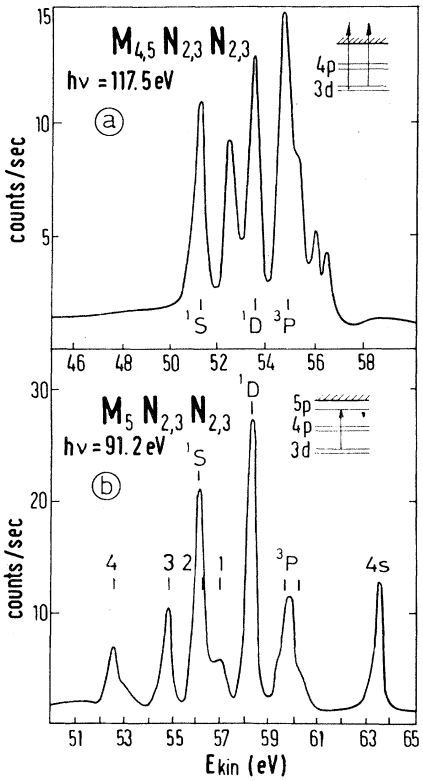

FIG. 3. *MNN* Auger spectra of Kr at two different photon energies: (a) above threshold of ionization of the $3d$ shell, and (b) at the energy for the $5p$ resonance excitation of the $3d_{5/2}$ shell. The peak labeled $4s$ is due to the direct ejection of electrons from the $4s$ shell.

the energy E_i injected into the system and the energies E_f of possible final-state configurations. These are, in our special case, $E_i = h\nu = 65$ eV for the resonance excitation from $4d^{10}5s^25p^6 \rightarrow 4d^95s^25p^6(^2D_{5/2})6p$ and $E_f = 29.5$ eV for producing the $5s^25p^4(^1S)6p$ configuration. The kinetic energy $E_{kin} = h\nu - E_f = 35.5$ eV for the ^1S peak falls at a 5.9-eV higher energy than the energy of the same peak when exciting far above threshold. [For the case of Xe^{++} we obtain $E_{kin} = E_B - E_f = 29.6$ eV with the binding energy[8] $E_B = 67.55$ eV of a $4d_{5/2}$ electron and[7] $E_f = 37.96$ eV for producing the $5s^25p^4(^1S)$ configuration.] In analogy, we calculate an energy difference of about 4.4 eV for the ^3P peaks. The energy difference of about 1.5 eV between these values indicates the different coupling of the $6p$ electron to the $5s^25p^4$ core. The electron energies $E_{kin} = h\nu - E_f$ are indicated in Figs. 2 and 3 for the resonance excitation. The energies are in good agreement with observed maxima. In contrast to the case of excitation above threshold, only one series is involved because of our selective excitation of the $4d_{5/2}$ state of the hole in Xe ($3d_{5/2}$ for Kr). This makes the interpretation of the spectra easier.

In addition to these main Auger lines, which we could identify after Moore, new peaks (1–4 in Figs. 2 and 3) are observed. These peaks probably result from a shake-up process accompanying the Auger transition. A shake-up process is generally described as a transition between two states as the result of a sudden perturbation. It is a transition conserving parity (see, e.g., Åberg[9]). For the series $5s^25p^4np$ of Xe^+ we estimate an energy separation of 3.3 eV between the $5p^46p$ and $5p^47p$ states by taking the optical data[7] from the $5s^25p^56p$ and $5s^25p^57p$ states of Cs, since the energies of the $5s^25p^47p$ states of Xe^+ are not listed in Moore's tables.[7] A shift of the different states of the configurations $5s^25p^46p$ of Xe^+ by 3.3 eV leads to the positions marked 1–4 in Fig. 2(b). In a similar way, we obtain an energy separation of the same order of magnitude between the $4s^24p^45p$ and $4s^24p^46p$ states of Kr using the data[7] for Rb (Fig. 3). In both cases the good coincidence with the measured spectrum supports the relevance of this interpretation. The shake-up processes apparently have a probability of the same order of magnitude as those without shake-up. Analogously Krause and Carlson[10] found that for Kr $M_{4,5}NN$ the double electron emission, as a special case of shake-up in the Auger process, competes strongly (~ 30%) with the single Auger process.

We also have measured the Auger spectrum after excitation to the second resonance state, $4d^95s^25p^6(^1D_{5/2})7p$ at 66.3 eV for Xe [Fig. 2(d)]. The marks in Fig. 2(d) indicate the kinetic energy of the electrons calculated by $E_{kin} = h\nu - E_f - \Delta E_{6p\rightarrow 7p} = 66.3\ \text{eV} - E_f - 3.3$ eV, with the energy E_f of the $5s^25p^46p$ final-state configuration and the energy difference ΔE between the states $5s^25p^46p$ and $5s^25p^47p$ as estimated before. The marks are in good agreement with the main structures. If we have no shake-down, the mean structures on the high-kinetic-energy side should correspond to Auger processes without shake-up retaining a $5s^25p^47p$ final-state configuration. This also supports the assumption that after excitation to the first resonance line as discussed above the dominant structures besides the single Auger lines originate mainly from $5s^25p^46p \rightarrow 5s^25p^47p$ shake-up transitions. Contributions from shake-up to higher excited states, as for example $8p$ or $9p$ for Xe, could not be identified.

The spectrum of Xe [Fig. 2(c)] taken at the energy for the $6p$ resonance excitation of the spin-orbit partner of the electrons from the $4d$ shell, 67 eV for the excitation from the N_4 subshell, shows nearly the identical structures of Fig. 2(b) with a 2-eV higher electron energy corresponding to the higher photon energy. Only the peak at about 38.5 eV in Fig. 2(b) which we cannot really explain at the moment does not appear in that spectrum.

An attempt to attribute any peaks to shake-up processes connected with the directly excited $5p$ or $5s$ electrons of Xe[11] ($4p$, $4s$ for Kr) is not justified, since we could show that these are about one order of magnitude weaker. For proving this we have chosen an excitation energy between two resonance states of the $4d$ excitation, namely 65.8 eV between the $6p$ and $7p$ states of Xe (Fig. 1) and 92.8 eV for the $3d$ excitation of Kr. The cross section for the production of a d hole is practically zero there and therefore no Auger lines are observed. But, of course, the $5s$ and $5p$ ($4s$, $4p$) lines of the direct excitation are excited with practically the same intensity as at the resonance energies. The amplitudes of the satellites are only about 10% of the amplitudes of the Auger lines in the spectra Figs. 2 and 3.

A source with tunable photon energy permits a selective excitation of the atomic shells. Using synchrotron radiation we were able to perform a detailed analysis of the mechanism of deexcitation of the Xe $4d$ and Kr $3d$ resonance excitations. After excitation to the $6p$ (Xe) and $5p$ (Kr) reso-

nance line the usual Auger multiplet structure is generally shifted by the relaxation energy of about 5 eV for Xe and Kr to higher kinetic energies. We could also show that there is nearly the same probability for the two atoms to decay by the single Auger transition and by a Auger transition accompanied by a shake-up process leading to $5s^25p^47p$ and $4s^24p^46p$ excited states in Xe and Kr. The comparison between the measured spectra and the energies obtained from optical data[7] demonstrates the relevance of core rearrangement and other correlation effects in Auger transitions.

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Observation of MeV Dissociative H_2^+ Ions Emerging from Very Thin Foils

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We present angle and energy distributions of H atoms emerging from thin carbon foils bombarded with MeV H_2^+ and H_3^+ beams. If the target is thin enough, a projectile electron can participate in the formation of a neutral atom at emergence through dissociation of a vibrationally excited H_2^+ molecule. This process is not observable with thicker targets when the formation of a neutral requires the capture of a target electron. The results suggest that electrons can be bound to the projectile inside the solid.

Recent studies performed in our laboratory[1] have given new information about the neutralization of MeV protons at emergence from solids. It has been shown that hydrogen species containing electrons (H, H_2^+, H_3^+) produce more neutral atoms in the beam transmitted through very thin carbon foils (transit times $< 2\times10^{-15}$ s) than incident protons of the same velocity.

However, this evidence for what we shall call, perhaps abusively, a transmission of bound electrons through a solid target does not describe the electron-proton system when moving *inside* the solid. The existence of a bound state has been questioned by Brandt[2] and recently considered by Cross.[3] Moreover it was shown in Ref. 1 that the angular distributions of H atoms in the transmitted beam from incident H_2^+ projectiles are compatible with the repulsion of randomly oriented H^+H pairs along the dissociative $2p\sigma_u$ state of the H_2^+ molecule. This observation was, however, restricted to foil thicknesses for which the neutral atoms emerge after capture of a target electron.

In order to know more about the transmission effect in which the incident-projectile electrons participate in the formation of emergent H atoms, we have studied the angular distributions of H atoms transmitted through very thin carbon foils bombarded with H_2^+ and H_3^+ ions. This investigation was pursued further by measuring the ener-