Oscillations in Coincident Molecular-Orbital Auger Spectra Emitted in 450-keV Kr^{+4} + Kr Collisions

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With use of coincidence techniques, oscillations in energy spectra of electrons produced in impact-parameter–controlled 450-keV Kr^{+4} + Kr collisions have been observed. The decay of an autoionizing state of the Kr–Kr quasimolecule is found to be the dominant electron emission mechanism. The superposition of the emission amplitudes for the incoming and the outgoing parts of the nuclear trajectory explains very well the oscillatory pattern.

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There has been increasing interest in the study of the continuum part of energy spectra of electrons produced in adiabatic heavy-ion–atom collisions.[1,2] While the discrete part of the spectra is rather well understood now, the mechanisms leading to the emission of electrons with a continuous spectrum are not yet clear. Especially a bandlike structure in the electron spectra produced in Kr+Kr collisions has been investigated in detail by measurements of singles spectra (without fixing the nuclear trajectory)[1,2] and by measurements of coincident spectra in collisions with large impact parameters.[3] The conclusions drawn from the experimental results indicate that the process of molecular autoionization [analogous to the emission of molecular-orbital (MO) x rays] is the origin of the observed structures.

The decay of an autoionizing state in a quasimolecule formed in an adiabatic collision can happen on the incoming and on the outgoing parts of the nuclear trajectory. The superposition of the two emission amplitudes should lead to observable structures in the electron spectra, provided the trajectory of the two colliding particles is fixed. In the present paper these oscillatory structures are reported in an electron–scattered-particle coincidence experiment with 450-keV Kr^{+4} ions impinging upon a Kr gas target. The oscillatory pattern also gives a crucial test of MO calculations.

The experiment was performed at the AMI accelerator[4] at the Fundamenteel Onderzoek der Materie–Instituut. 450-keV Kr^{+4} ions were collimated by three slits to an angular divergence smaller than 1.4 mrad before they hit the Kr gas target. Since the experimental setup has been described extensively,[2] only the main characteristics of the target, the particle-detection system, and the electron energy analyzer are given here.

The Kr gas target which was produced by flow through a capillary had a pressure below 8×10^{-5} Torr in the scattering center. The scattered primary ions were detected by an annular surface barrier detector whose distance from the target could be varied continuously so that an angular region of $1.8^\circ \leq \theta \leq 14^\circ$ in the laboratory system could be covered. The scattering angle was defined by an annular slit of 1 mm width and 20 mm diameter mounted in front of the detector.

The energy of the electrons produced in the collisions was determined by a cylindrical mirror analyzer which has been constructed based on the calculations by Van Hoof.[5] The angle of observation is $\theta = 141^\circ$ (thus in the backward direction) and the energy resolution of the analyzer is $\Delta E_e/E_e = 2.2\%$ [full width at half maximum (FWHM)]. The total solid angle provided by the analyzer and three channeltrons for electron detection is 0.12 sr. The absolute efficiency of analyzer and detection system was determined with use of the absolute electron emission cross sections in 250- and 300-keV Ar^+ + Ar collisions[6] and in 800-keV Ne^{3+} + Ne collisions.[2] The influence of a possible scattering angle dependence of the effective target volume on the data is negligible. This was found from a measurement of the θ dependence of the production probability of a part of the L Auger electron spectrum in 600-keV Ar^{3+} + Ar collisions and a comparison with existing experimental data.[7]

Coincidences between electrons and scattered particles at a fixed electron energy were measured with the standard fast-slow techniques. Because of the small signal-to-noise ratio from the particle detector special care was taken to achieve a signal processing free of any distortion. A time analyzer with a range of 10 μsec had to be used for a proper determination of the number of

true coincidences since the time spectrum was strongly influenced by (irreproducible) plasma oscillations in the ion source with frequencies ranging from 100 kHz to several MHz. The coincidence time resolution finally reached was about 200 nsec resulting in a true-to-random ratio ranging from 2 to 5.

The measured electron spectra are shown in Fig. 1. Plotted is the probability $P(E_e,\theta)$ for the emission of an electron with energy E_e in a collision with a scattering angle θ, as a function of E_e. $P(E_e,\theta)$ and E_e are transformed into the c.m. system using the familiar expressions for the Doppler effect. While in singles electron spectra only a broad band for $100 \lesssim E_e \lesssim 300$ eV has been observed,[2] the coincident spectra exhibit a distinctive oscillatory structure. With increasing scattering angle this structure shifts to higher energies and the amplitudes of the oscillations strongly increase. Beyond a certain (θ-dependent) electron energy the structure disappears and a pure exponential decay with E_e is observed.

It has been argued[1,2] that the broad band in the singles spectra is due to the autoionizing decay of a vacancy in a level of the Kr_2 quasimolecule which correlates to the $n=4$ state in the united atom. Since autoionization can occur on the incoming and on the outgoing parts of the trajectory, the superposition of the two corresponding amplitudes should result in oscillations in the electron spectra, provided the trajectory is experimentally fixed and the coherence is preserved. The structure of these oscillations has been investigated in great detail theoretically and experimentally for molecular autoionization of outer shell levels at low projectile energies.[8] An equivalent formulation which is easily applicable to higher energy collisions and inner shells as in the present case has been developed by Fritsch and Wille[9] for the emission of MO x rays. On replacement of the matrix element for photon emission by the corresponding one for autoionization, the emission probability of an electron with energy E_e at a projectile velocity v and a distance of closest approach R_0 is given by

$$P(E_e,R_0)=a_1\,\mathrm{Ai}^2(x)+a_2\,\mathrm{Ai}'^2(x), \tag{1}$$

where

$$x=[E_e-\Delta E(R_0)]\beta^{-1/3}(\hbar v)^{-2/3}, \tag{2}$$

and

$$\Delta E(R)=\Delta E(R=0)-\beta R^2. \tag{3}$$

In Eq. (1), where Ai and Ai′ denote the Airy function and its derivative, respectively, it is assumed that R_0 is large compared to half the distance of closest approach in a head-on collision. The energy difference $\Delta E(R)$ between the two molecular orbitals under consideration is parametrized according to Eq. (3) by a quadratic R dependence[8] with two free parameters $\Delta E(R=0)$ and β. Under the reasonable assumption that the high-

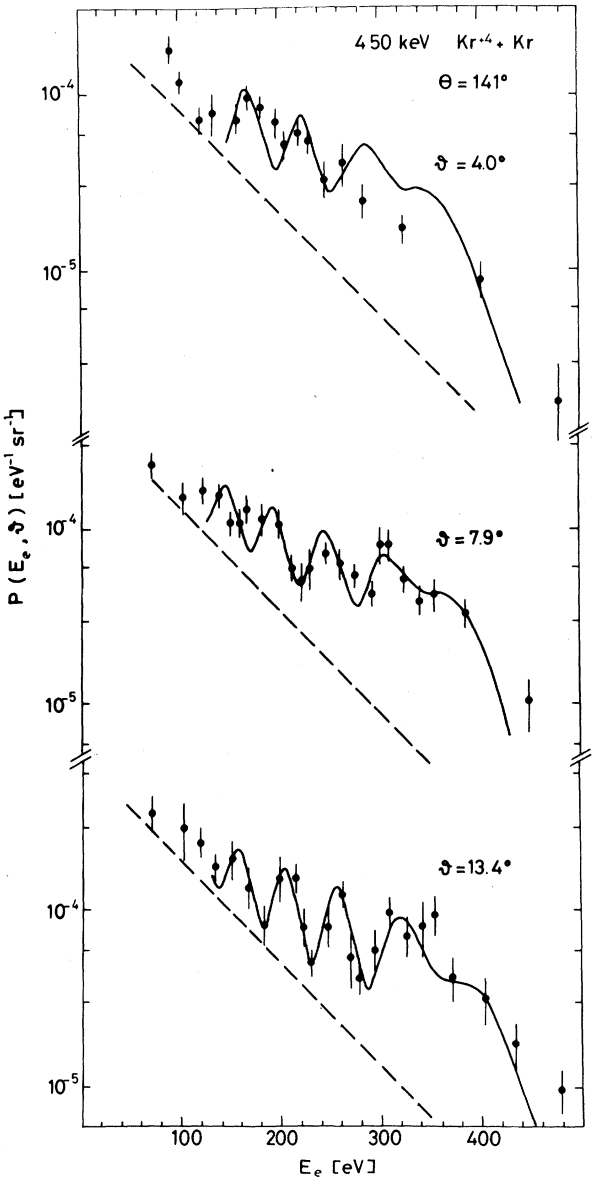

FIG. 1. Electron emission probability as a function of the electron energy E_e in 450-keV Kr^{+4} + Kr collisions at 4.0°, 7.9°, and 13.4° laboratory scattering angles under 141° observation angle. The solid lines give the calculated emission probability from a superposition of the background of δ electrons (dashed lines) and of the molecular autoionization [Eqs. (1) – (3)].

er-lying orbital is only weakly bound, Eq. (3) gives the R dependence of the binding energy of the autoionizing orbital. At a given collision velocity v, the coefficients a_1 and a_2 depend on the properties of the individual collision system.

Equation (1) predicts an oscillatory structure of the electron spectrum with a broad peak at an electron energy comparable to the binding energy at the turning point. This peak has already been observed experimentally at a lower projectile energy.[3]

In order to apply Eq. (1) to the data in Fig. 1, the inherent "background" of δ electrons with an exponential energy spectrum has to be subtracted. From experimental emission cross sections in 400-keV Kr^{+4}+Kr collisions[2] it is found that for electron energies $E_e \gtrsim 400$ eV the spectrum is well described by an exponential $\exp[-E_e/(74\ \text{eV})]$. This part of the spectrum is caused by electron emission from highly promoted MO into the continuum at large internuclear distances,[2] and thus a change of the spectral shape for the small impact parameter collisions of the present experiment is most unlikely.[10] The absolute values of this contribution to the electron spectra were fixed from the lowest-lying experimental point for every spectrum separately and are given by the dashed lines in Fig. 1.

By application of Eq. (2) to the remaining $P(E_e, \theta)$ data at $\theta = 13.4°$ (this corresponds to $R_0 = 0.21$ a.u. for the appropriate[11] screened Coulomb potential), the curvature β and the energy difference $\Delta E(0.21\ \text{a.u.})$ were obtained from the positions of zeros and maxima of $\text{Ai}(x)$ and $\text{Ai}'(x)$[12] as $\beta = 11.21$ a.u. and $\Delta E(0.21\ \text{a.u.}) = 14.89$ a.u. The absolute values of $P(E_e, \theta)$ at maxima and minima gave for the coefficients in Eq. (1) $a_1 = 1.2 \times 10^{-4}$ and $a_2 = 1.65 \times 10^{-4}$. The final result for the electron emission probability $P(E_e, \theta)$ is given by the incoherent superposition of the background contribution and the molecular autoionization (solid line in Fig. 1).

Via Eq. (3), the energy difference $\Delta E(R)$ at $R_0 = 0.28$ and $R_0 = 0.38$ a.u. (corresponding to $\theta = 7.9°$ and $\theta = 4°$, respectively) is given by $\Delta E(0.28\ \text{a.u.}) = 14.5$ and $\Delta E(0.38\ \text{a.u.}) = 13.8$ a.u. Thereby, the position and the wavelength of the oscillatory structure are fixed [cf. Eqs. (1) and (2)], and only a_1 and a_2 are left as free parameters. Proceeding as before, we obtained $a_1 = 1.35 \times 10^{-4}$ and $a_2 = 1.2 \times 10^{-4}$ for $R_0 = 0.28$ a.u. and $a_1 = a_2 = 9 \times 10^{-5}$ for $R_0 = 0.38$ a.u. The solid lines again give the thus calculated emission probability $P(E_e, \theta)$.

The energy difference $\Delta E(R)$ calculated from Eq. (3) is plotted versus R in Fig. 2 and is compared to the adiabatic $4(\frac{3}{2})u$ level (which correlates to the $4p$ orbital in the separated system) from relativistic self-consistent-field calculations for the Kr_2^{4+} molecule from Fricke and Sepp.[13] The agreement between these calculations and the experimental results is very good. At larger R where the simple parabolic expansion with a constant curvature β can no longer be applied, the theoretical data are also in very good agreement with the results extracted by Gordeev *et al.* from singles spectra[2,13] which are not very sensitive to small-impact-parameter collisions. Thus, the energy difference extracted from the oscillatory structure strongly indicates that a vacancy in the $4(\frac{3}{2})u$ level (provided by initial $4p$ vacancies in the projectile) forms the autoionizing state.

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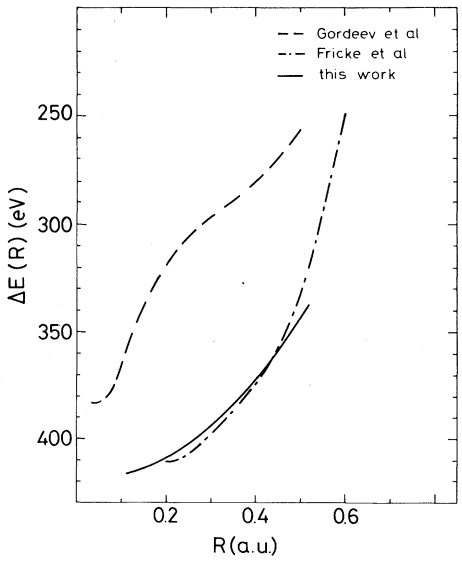

FIG. 2. Energy difference $\Delta E(R)$ for the Kr^{+4} + Kr system extracted from singles spectra (Ref. 2) and from the present coincidence spectra. The dash-dotted line gives the adiabatic $4(\frac{3}{2})u$ orbital from relativistic self-consistent-field calculations (Ref. 13).

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