Coincidence studies of quasimolecular electron emission in 700-keV Ar^{2+}-Kr collisions

R. Shanker
Department of Physics, Banaras Hindu University, Varanasi-221 005, India

U. Werner, R. Bilau-Faust,* and R. Hippler
Fakultät für Physik, Universität Bielefeld, D-4800 Bielefeld, Federal Republic of Germany

U. Wille
Bereich Kern- und Strahlenphysik, Hahn-Meitner-Institut für Kernforschung Berlin G.m.b.H., Postfach 390128, D-1000 Berlin 39, Federal Republic of Germany

(Received 21 February 1989)

Continuum electron emission following 700-keV Ar^{2+}-Kr collisions has been investigated by detecting electrons in coincidence with projectile ions scattered through laboratory angles between 5° and 20°. From the analysis of the experimental data in terms of quasimolecular Auger-electron emission, molecular-orbital binding energies at small internuclear distances as well as collision-broadening parameters have been inferred. The results indicate that the electron emission is dominated by the decay of $4s\sigma$ and $4p\pi$ vacancies in the highly charged Ar-Kr quasimolecule. The analysis also reveals the presence of an exponentially decreasing high-energy tail of the spectra, which cannot be ascribed to quasimolecular Auger electrons and which has not been taken into account in previous analyses.

I. INTRODUCTION

In slow atomic collisions, i.e., in collisions where the projectile velocity is much smaller than the (classical) orbital velocity of bound electrons, the collision partners transiently form a quasimolecule (see, e.g., Wille and Hippler,[1] and references therein). Vacancies present in outer shells of projectile or target prior to the collision are transferred during the course of the collision from "demoted" quasimolecular orbitals via couplings with more deeply bound states into inner quasimolecular orbitals. *After* the collision these vacancies eventually end up in inner atomic shells, where they decay via emission of characteristic x rays or Auger electrons. However, a certain fraction of vacancies already decays *during* the collision, thereby giving rise to the emission of a continuous spectrum of quasimolecular electrons or x rays.[2] In this paper, we report on an experimental study of quasimolecular electrons ejected during 700-keV Ar^{2+}-Kr collisions. Making use of the scattered-projectile–ejected-electron coincidence technique, electron spectra for fixed ion trajectories have been recorded. From the theoretical analysis of these spectra, we have inferred information on molecular-orbital binding energies and collision-broadening parameters, particularly in the range of small internuclear distances. In this respect, our work supplements and extends previous (noncoincident) studies of Afrosimov *et al.*[3]

II. THEORY

Theoretically, the doubly-differential cross section $d^2\sigma/dE_e db$ (differential in electron kinetic energy E_e and ion impact parameter b) for electron emission due to Auger decay of vacancies in a specific (demoted) quasimolecular orbital may be expressed within the simple Airy approximation as[4,5]

$$\frac{d^2\sigma}{dE_e db}=4nb\Gamma_a\alpha^{-2/3}\mathrm{Ai}^2(\alpha^{-1/3}[E_e-E(R_0)+\tfrac{1}{2}i\Gamma_a]) , \tag{1}$$

where $E(R_0)$ is the Auger transition energy at the minimum value R_0 of the internuclear separation $R=R(t)$, Γ_a is the Auger level width,

$$\alpha=\frac{1}{2}\frac{dE}{dR}\frac{d^2R}{dt^2}\bigg|_{R=R_0} , \tag{2}$$

and n is the number of vacancies initially present in the quasimolecular orbital.

Beyond the classically allowed maximum value of the electron kinetic energy [which for Auger decay of vacancies in a demoted orbital is given by $E(R_0)$], the spectrum according to Eq. (1) exhibits a tail [collision broadening (CB)] characterized by the width[5]

$$\Gamma_{\mathrm{CB}}=0.42\alpha^{1/3} . \tag{3}$$

The asymptotic form of Eq. (1) for $E_e\rightarrow\infty$ is

$$\frac{d^2\sigma}{dE_e db}\sim\exp\{-\tfrac{4}{3}\alpha^{-1/2}[E_e-E_e(R_0)]^{3/2}\} , \tag{4}$$

i.e., the differential cross section decreases faster than exponentially.

III. EXPERIMENTAL ARRANGEMENT

Our experiments were performed at the 350-kV ion accelerator at the Universität Bielefeld, West Germany (e.g., Luz *et al.*[6] and Bilau *et al.*[7]). A well collimated beam of 700-keV Ar^{2+} ions was injected into a dilute-krypton-gas target (typical gas pressure 10^{-3} mbar) to ensure single-collision conditions. Scattered projectiles were detected by a set of four cooled surface-barrier detectors mounted on an annular ring; a given scattering angle was selected by moving this annular ring along the beam direction. Ejected electrons were observed at 90° with respect to the ion beam by a hemispherical energy analyzer (energy resolution $\Delta E_e/E_e=0.035$) and detected with an open electron multiplier. The overall efficiency of the electron analyzer-detector combination was investigated by comparing a measured electron spectrum from 300-keV H^+-Ar collisions with data compiled by Rudd *et al.*[8] The overall efficiency thus determined increases slightly smaller than linearly with electron energy E_e, in fair agreement with the predicted transmission curve for this type of electron analyzer.[9] The measured spectra have been corrected accordingly. Pulses from electron and ion detectors served, after suitable amplification, as start and stop inputs, respectively, for a time-to-amplitude converter (TAC). Suitable time windows were chosen for the TAC output to allow simultaneous recording of "true-plus-random" and "random" coincidences. Coincident and noncoincident electron spectra have been recorded simultaneously; data acquisition, control, and analysis were carried out using a PDP 11/20 computer.

IV. RESULTS AND DISCUSSION

Our experimental results are shown in Fig. 1, where the differential cross sections (DCS) for electron ejection in 700-keV Ar^{2+}-Kr collisions are plotted versus the energy of ejected electrons. Five electron spectra are displayed, of which the total spectrum was measured without coincidence requirement. This total spectrum was obtained from the measured number of ejected electrons N_e, which is related to the doubly-differential cross section $d^2\sigma/dE_e d\Omega_e$ via

$$\frac{d^2\sigma}{dE_e d\Omega_e}=\frac{N_e}{\Omega_e \epsilon_e \Delta E_e N_i N_a L}\ , \tag{5}$$

where Ω_e and ϵ_e are solid angle and efficiency of the electron spectrometer, respectively, ΔE_e is the energy resolution, N_a and N_i are target density and number of incident projectiles, respectively, and L is the observation length. The other four spectra constitute a triply-differential cross section $d^3\sigma/dE_e d\Omega_e d\Omega_i$; they were obtained in coincidence with projectiles scattered through scattering angles of 5°, 10°, 14°, and 20°, respectively. This corresponds to ion trajectories with impact parameters b ranging from 0.058 to 0.18 a.u.; the corresponding distances of closest approach R_0 vary between 0.072 and 0.19 a.u. Transformation of scattering angles into impact parameters and distances of closest approach was achieved with the help of a Molière potential (e.g., Shanker *et al.*[10]).

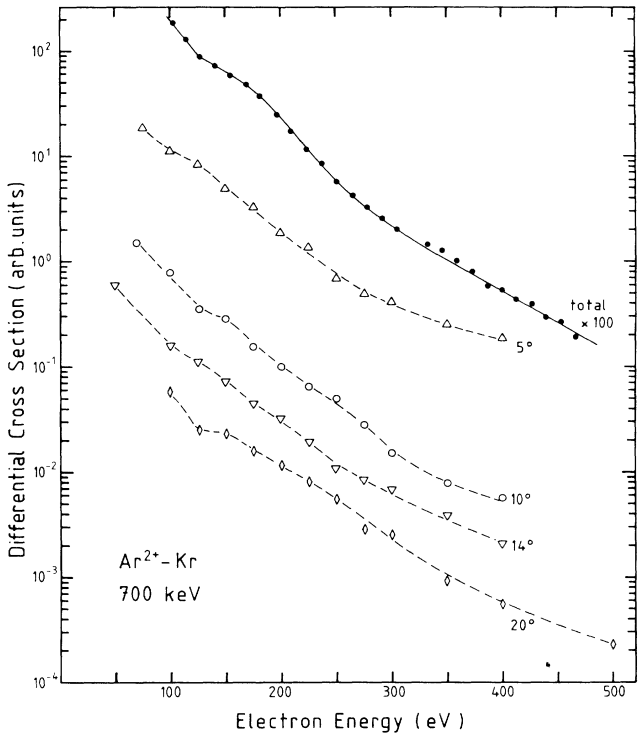

FIG. 1. Differential cross sections for electron ejection in 700-keV Ar^{2+}-Kr collisions vs electron energy. The total (noncoincident) electron spectrum (●) and the electron spectra measured in coincidence with projectiles scattered through angles $\theta_i=5°$ (△), 10° (○), 14° (▽), and 20° (◇) are displayed (dashed and solid curves serve to guide the eye only).

The triply-differential cross sections (TDCS's) were extracted from the measured number of coincidence counts N_c by using

$$\frac{d^3\sigma}{dE_e d\Omega_e d\Omega_i}=\frac{N_c}{\Omega_i \epsilon_i \Omega_e \epsilon_e \Delta E_e N_i N_a L}\ , \tag{6}$$

where Ω_i and ϵ_i are solid angle and efficiency of the ion detector, respectively. Based on the counting statistics, the relative accuracy of each of the coincident data points is ±10%.

Common features of all five spectra are (i) an exponential decrease with increasing electron energy of the DCS below about 100 eV, (ii) the appearance of a shoulderlike structure between electron energies of 100 and 300 eV, and (iii) an exponential tail above about 300 eV. The broadband structure between 100 and 300 eV is believed to be due to the emission of quasimolecular Auger electrons ejected *during* the collision. The exponential tail above 300 eV can be only partly ascribed to this source since the high-energy tail of the quasimolecular Auger-electron spectrum is expected to depart from a strictly exponential behavior [cf. e.g., Eq. (4)]. The parts of the spectra below 100 eV are expected to result from direct ionization of outer-shell electrons during the collision. This contribution to the measured DCS is known to ex-

hibit a single exponential decrease over the whole range of electron energies.[11]

In order to extract the quasimolecular contribution, we have fitted, in a first attempt, the coincident electron spectra by a sum of a single exponential plus the square of an Airy function. Also, we have neglected the imaginary part in the argument of the Airy function. Using realistic estimates[3] for the quasimolecular transition probabilities of $\approx 5\times 10^{15}$/s we expect an autoionizing level width $\Gamma_a \approx 3$ eV; this value is certainly small compared to electron energies of the order of a few hundred eV and accordingly can be neglected. It turned out that such a least-squares fit is unable to satisfactorily reproduce the high-energy part of the electron spectra above 300 eV; the almost pure exponential decrease of the TDCS in this range cannot be accomodated in the high-energy behavior of the Airy function. The origin of the additional contribution to the TDCS that must be assumed to be present in the high-energy range is unknown. It could be caused by direct or quasimolecular electron ejection from more deeply bound inner shells, and was, in our analysis, accounted for by including a second exponential into the fitting routine. Hence we have fitted the measured TDCS according to

$$\frac{d^3\sigma}{dE_e d\Omega_e d\Omega_i} = c_1 \exp(-c_2 E_e) + c_3 \mathrm{Ai}^2(c_4(E_e - c_5)) + c_6 \exp(-c_7 E_e) , \quad (7)$$

where c_1 to c_7 are independent parameters. A typical result of such a fit is displayed in Fig. 2. It shows the measured TDCS for $\theta_i = 20°$ and its decomposition into the contributions corresponding to the three terms of Eq. (7). From the fitting procedure we find that the low-energy part of all four TDCS's is given by a single exponential; the slope of this exponential amounts to $c_2 \approx 0.03$/eV for all spectra. A theoretical explanation for this specific value is obviously beyond the scope of the present paper.

In Fig. 3 we display our results for the transition energies $E(R_0)$ as obtained from the fitted values of the parameter c_5 in Eq. (7). Our values for $E(R_0)$ provide a fairly smooth continuation of the (noncoincident) results of Afrosimov *et al.*[3] into the range of small internuclear distances R_0. Also shown in Fig. 3 are selected diabatic molecular-orbital energy curves as obtained from pertinent adiabatic curves calculated within the variable-screening model.[12] We may identify the transition energies with individual molecular-orbital binding energies if we assume the outer-shell orbitals involved in the Auger transition to have binding energies negligible in comparison with the binding energy of the demoted orbital carrying the initial vacancy.

The calculated energy curves of Fig. 3 refer to the Ar^{3+}-Kr^{3+} and Ar^{4+}-Kr^{4+} systems, i.e., to a total charge of the quasimolecular system of 6+ and 8+, respectively. In calculating energy curves for *charge-symmetric* collision systems, we have assumed that at the small internuclear distances relevant for the present case the collision system has completely "forgotten" the initial distribution of electrons about projectile ion and target atom. It is seen from Fig. 3 that the experimental transition energies are in good agreement with both the $4s\sigma$ and $4p\pi$ molecular-orbital binding energies corresponding to a total charge 8+. The comparison with the 6+ case illustrates the fairly strong dependence of the binding energies on the charge state; in particular, it shows that the $4s\sigma$ and $4p\pi$ orbitals corresponding to the initial charge state 2+ have binding energies much smaller than

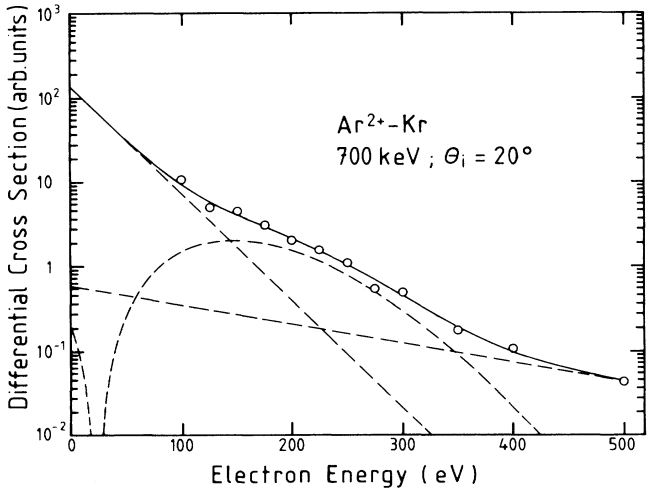

FIG. 2. Triply-differential cross section for electron ejection in 700-keV Ar^{2+}-Kr collisions and for a scattering angle $\theta_i = 20°$ vs electron energy (○). The solid line is a least-squares fit according to Eq. (7); dashed lines represent the decomposition of the fitted spectrum into the contributions of the three terms of Eq. (7).

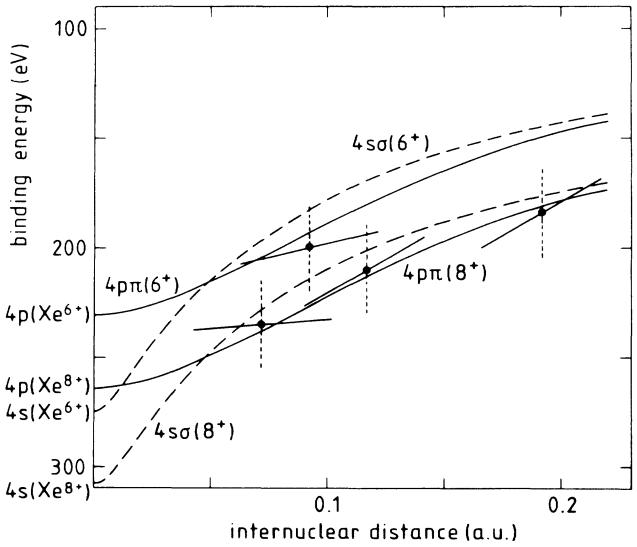

FIG. 3. Selected (diabatic) molecular-orbital energy curves for the $(Ar\text{-}Kr)^{n+}$ ($n = 6, 8$) collision systems as calculated from the variable-screening model (Ref. 12). Also shown are present experimental results (●) for $E(R_0)$, and for the slope $dE/dR|_{R_0}$ [solid bars attached to the $E(R_0)$ points].

the experimental energies. Molecular orbitals other than $4s\sigma$ and $4p\pi$ cannot provide any satisfactory explanation of the experimental data: the orbitals correlating to the united-atom $4d$ and $4f$ shells are *promoted* orbitals, while the orbitals correlating to the united-atom $5s$ to $5g$ shells have much too small binding energies. The occurrence of charge states as high as $8+$ in the quasimolecular Ar-Kr system at small internuclear distances appears plausible in view of the large amount of outer-shell excitation and ionization that is known to take place in the initial phase of heavy ion-atom collisions (in principle, the dynamical charge state of the collision system depends on the impact parameter; allowing, in the present analysis, for such a dependence will, however, hardly change the results and conclusions).[13]

As the $4s\sigma$ and $4p\pi$ binding energies for charge state $8+$ agree almost equally well with the experimental transition energies, we cannot assign a single quasimolecular orbital to the observed Auger-electron spectra. We mention that, due to many low-lying separated-atom levels, the diabatic correlations[1] of the $4s\sigma$ and $4p\pi$ orbitals cannot be uniquely decided. Accordingly, we are not able to estimate the number of vacancies initially present in these molecular orbitals from the distribution of vacancies over the separate-atom orbitals. A slight indication for a large number of vacancies to be present in the $4s\sigma$ orbital can be derived from the fact that this orbital behaves, for small internuclear distances, more diabatic than the $4p\pi$ orbital (cf. the adiabatic Ar-Kr correlation diagram shown in Ref. 7).

Apart from the experimental transition energies, we have displayed in Fig. 3 values for the slope $dE/dR|_{R_0}$ as extracted from the fitted values of the parameter c_4 [cf. Eq. (7)] by identifying c_4 with $\alpha^{-1/3}$ and using Eq. (2). It is seen that the experimental values for the slope follow roughly the behavior of the $4p\pi$ curve. Experimental values for the collision broadening Γ_{CB} have been determined from the c_4 values via Eq. (3) and are displayed in Fig. 4. Also shown in this figure are theoretical values for Γ_{CB} calculated via Eq. (3) from the diabatic $4s\sigma$ and $4p\pi$ energy curves of Fig. 3. The discrepancies between the experimental and theoretical values for Γ_{CB} are, of course, linked to the discrepancies between the experimental and theoretical slopes $dE/dR|_{R_0}$ in Fig. 3.

A rough comparison of our results for Γ_{CB} to the (noncoincident) results of Shergin *et al.*[14] for 50–500-keV Kr^+-Ar collisions can be made by extrapolating the latter results to 700-keV Ar-Kr collisions assuming $\Gamma_{CB} \sim v_p^{2/3}$. Such a dependence has been predicted theoretically[15] and was confirmed by the data of Shergin *et al.*[14] Extrapolation of the latter results to our case gives $\Gamma_{CB} \approx 57$ eV, which is a factor of 1.5–2 larger than our values shown in Fig. 4. Shergin *et al.*[14] have noted that their results are generally larger by a factor of about 2 than theoretical predictions for Γ_{CB}; a similar discrepancy was observed by Shanker *et al.*[16] for Kr-Xe collisions. A possible reason for all these discrepancies can be found in the fact that Shergin *et al.*[14] and Shanker *et al.*[16] have derived Γ_{CB} from the high-energy tail of the measured electron spectra. This high-energy part was observed to fall off exponentially, whereas the electron spectrum according to Eq. (4) decreases *faster* than exponentially. According to the present study, the high-energy tail is in fact dominated by an exponentially decreasing background contribution (of admittedly unknown origin), which must be properly taken into account in the analysis.

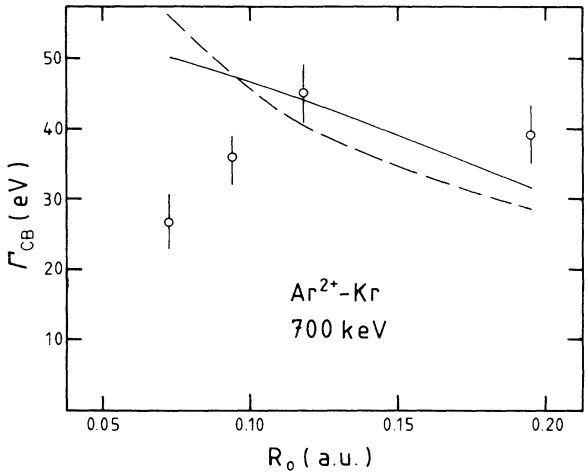

FIG. 4. Collision broadening $\Gamma_{CB}(\circ)$ vs distance of closest approach R_0 in 700-keV Ar^{2+}-Kr collisions. Also shown are the theoretical predictions for the $4s\sigma$ (dashed line) and $4p\pi$ (solid line) molecular orbitals with charge state $8+$ of the quasimolecular Ar-Kr system.

V. CONCLUSIONS

The coincidence study of continuous electron emission in slow Ar-Kr collisions has enabled us to investigate quasimolecular Auger transition energies particularly at small internuclear distances. The analysis of our data has revealed the importance of a careful decomposition of the measured electron spectra into the quasimolecular part and the background contribution. In particular, an exponentially decreasing high-energy tail, which cannot be attributed to quasimolecular Auger electrons, has been identified in the spectra and has been taken into account in the analysis of the data. From a comparison with realistically calculated molecular-orbital energy curves, we have inferred that Auger electron emission in the quasimolecular Ar-Kr system is dominated by $4s\sigma$ and/or $4p\pi$ vacancy decay in the highly charged system.

ACKNOWLEDGMENTS

We acknowledge helpful discussions with Professor Dr. H. O. Lutz. One of us (R.S.) gratefully acknowledges the comments and valuable suggestions received from Professor D. K. Rai and Professor S. N. Thakur. Part of this work was supported by the Deutsche Forschungsgemeinschaft.

*Permanent address: Max-Planck-Institut für Plasmaphysik, Boltzmannstrasse 2, D-8046 Garching, Federal Republic of Germany.

[1]U. Wille and R. Hippler, Phys. Rep. **132**, 129 (1986).

[2]R. Anholt, Rev. Mod. Phys. **57**, 995 (1985).

[3]V. V. Afrosimov, Yu. S. Gordeev, A. N. Zinov'ev, D. Kh. Rasulov, and A. P. Shergin, Pis'ma Zh. Eksp. Teor. Fiz. **24**, 33 (1976) [JETP Lett. **24**, 28 (1976)]; V. V. Afrosimov, Yu. S. Gordeev, A. N. Zinov'ev, A. I. Krotkov, and A. P. Shergin, in *Abstracts of the Tenth International Conference on the Physics of Electronic and Atomic Collisions, Paris, 1977,* edited by M. Barat and J. Reinhardt (Commissariat à l'Energie Atomique, Paris, 1977), p. 924.

[4]A. Z. Devdariani, V. N. Ostrovskii, and Yu. N. Sebyakin, Zh. Eksp. Teor. Fiz. **73**, 412 (1977) [Sov. Phys.—JETP **46**, 215 (1977)].

[5]V. V. Afrosimov, G. G. Meskhi, N. Tsarev, A. P. Shergin, Zh. Eksp. Teor. Fiz. **84**, 454 (1983) [Sov. Phys.—JETP **57**, 263 (1983)].

[6]N. Luz, S. Sackmann, and H. O. Lutz, J. Phys. B **12**, 1973 (1979).

[7]R. Bilau, W. R. McMurray, U. Wille, R. Shanker, R. Hippler, and H. O. Lutz, Z. Phys. D **1**, 39 (1986).

[8]M. E. Rudd, L.H. Toburen, and N. Stolterfoht, At. Data Nucl. Data Tables **23**, 405 (1979).

[9]H. D. Polaschegg, Appl. Phys. **4**, 63 (1974).

[10]R. Shanker, R. Hippler, U. Wille, and H. O. Lutz, J. Phys. B **15**, 2041 (1982).

[11]P. H. Woerlee, Yu. S. Gordeev, H. W. deWaard, and F. W. Saris, J. Phys. B **14**, 527 (1981).

[12]J. Eichler and U. Wille, Phys. Rev. A **11**, 1973 (1975).

[13]R. Shanker, R. Bilau, R. Hippler, U. Wille, and H. O. Lutz, J. Phys. B **14**, 997 (1981).

[14]A. P. Shergin, R. Mann, and H. F. Beyer, Z. Phys. A **302**, 191 (1981).

[15]W. Fritsch and U. Wille, J. Phys. B **12**, L335 (1979).

[16]R. Shanker, R. Hippler, R. Bilau-Faust, and H. O. Lutz, Phys. Lett. A **123**, 167 (1987).