Auger spectroscopy of quasimolecules

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The ionization occurring in atomic collisions at keV energies, and due to Auger transitions in the quasimolecule produced in the collision, is investigated. The Auger transitions in quasimolecules is studied for the first time by recording electrons by coincidences with ions scattered into a prescribed angle. In this way it is possible to choose collisions with a fixed impact parameter. The energy spectra of electrons produced in the decay of autoionizing states of quasimolecules in collisions between the ions and oxygen, neon, or krypton atoms are investigated in the 5–50 keV range and for impact parameters between 0.1 and 1. Quantitative spectroscopy of quasimolecules is attained in the investigations, and experimental data are obtained regarding the energies and widths of the quasimolecular levels. It is also found that owing to the existence of vacant dropping orbitals the Auger transitions in quasimolecules possess appreciable cross sections serve in atomic collisions as one of the main mechanisms of formation of electrons with a continuous energy distribution.

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I. INTRODUCTION

A new recent trend in the physics of atomic collisions is spectroscopy of the quasimolecule, which is a short-lived system made up of colliding atomic particles as they come close together and decay in flight. The importance of these investigations is due to the fact that electronic transitions between the terms of the quasimolecule determine to a considerable degree the picture and the values of the cross sections of the inelastic processes occurring in atomic collisions.

A quasimolecule is characterized by a set of electron states whose energies vary with the internuclear distance. When the nuclei come very close together, the quasimolecule is as a rule in one of the continuum states, i.e., emission of electrons or photons from the quasimolecule becomes possible via Auger or radiative transitions. The energy distribution of the emitted electrons and photons reflects the behavior of the energy and of the width of such an autoionizing level when the internuclear distance is changed. An analysis of these energy distributions can thus be used in principle for the spectroscopy of the quasimolecule.

The main cause of the onset of an autoionizing state of a quasimolecule is the formation of internal vacancies. In some cases, when the particles come sufficiently close together, the formation of vacancies has a probability close to unity. Among such cases are collisions of multiply charged ions with atoms, where the vacant levels have been produced beforehand, and also all collisions in which the vacancies are produced, upon approach of the particles, as a result of lowering of the molecular orbitals produced from the unfilled outer levels of the interacting ions or atoms.

The radiation produced upon decay of a vacancy in a quasimolecule was first investigated by Saris.[1] By now, radiative transitions in a quasimolecule have been quite extensively investigated.[2] However, the x-ray spectroscopy can be used only for deep levels. For shallower inner shells with electron-binding energies up to several keV, the principal role in the vacancy decay is played by Auger transitions.

The importance of extracting information on the energies and widths of quasimolecular levels from the energy spectra of electrons released in atom-atom collisions was first demonstrated in our earlier paper.[3] To observe the Auger transitions in a quasimolecule, we used a method based on measuring the energy spectra of the electrons produced in collisions with all possible impact parameters.

In the present investigation we recorded the electrons for coincidence with the ions scattered through a given angle. Since the scattering angle is determined by the impact parameter, this method makes it possible to measure spectra at fixed impact parameters, thereby increasing substantially the accuracy with which the energy and level widths of the quasimolecule are determined. In a number of cases this turns out to be the decisive factor that permits observation and interpretation of quasimolecular Auger spectra.

II. EXPERIMENTAL METHODS

The experimental setup for the investigations by the "electron-scattered ion" coincidence method is shown in Fig. 1. A beam of ions of energy E_0 is directed into the collision chamber TC filled with the target gas. The electrons produced upon interaction of the beam ions with the target-gas atoms were energy-analyzed in a cylindrical electrostatic analyzer A_e and recorded by detector D_e. The ions scattered at a fixed angle ϑ were recorded by detector D_i. The pulses in both detectors were fed to a delayed-coincidence circuit CC.

To study the Auger transitions in a quasimolecule it is necessary to bring the particles close to each other to within ~ 0.1 Å, corresponding to the dimensions of the inner electron shells. At keV collision-energies such distances are reached at impinging-ion scattering angles amounting to several degrees. The construction of the setup has made it possible to vary the angle ϑ in the range 0–18°.

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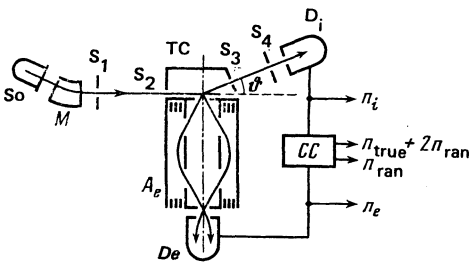

FIG. 1. Diagram of setup for the investigation of the energy spectra of electrons produced in atomic collisions with fixed impact parameters. *So*) Ion source; *M*) magnetic mass monochromatic; S_1 and S_2, S_3 and S_4) slits of the collimators of the first beam of ions and of the ions scattered through an angle ϑ; *TC*) target chamber; A_e) electron analyzer; D_e and D_i) electron and ion detectors; *CC*) coincidence circuit. n_e, n_i, n_{true}, and n_{ran} are respectively intensities of the recorded electrons, ions and true and random coincidences.

For an experiment for "electron-scattered ion" coincidence, a typical feature is that the intensity of the random coincidences n_{ran} exceeds the intensity of the true coincidence n_{true}. This is based on the fact that electrons having the same energy can be produced in collisions with different impact parameters, and also on the fact that the angular distribution of the released electrons is close to isotropic and the fraction of electrons separated by the analyzer aperture is quite small. For the case $n_{ran} \gg n_{true}$, the relative error δ is given by the expression

$$\delta^2 \sim \tau_r / t g_{ie} \Delta\Omega_i \varepsilon_i \Delta E_e \Delta\Omega_e \eta_e \varepsilon_e, \quad (1)$$

where τ_r is the resolution time of the coincidence circuit; t is the exposure time; g_{ie} is the degree of overlap of the volumes visible by the collimators of the ion and electron channels; $\Delta\Omega_i$ and $\Delta\Omega_e$ are the solid angles bounded by the collimators of the ions and electrons; ΔE_e is the width of the instrumental function of the electron energy analyzer; η_e is the transparency of the electron analyzer (the transparency of the ion channel is ~ 1); ε_i and ε_e are the efficiencies of recording the ions and electrons. It follows from (1) that the error is a minimum if the resolution time is short and the degree of overlap of the volumes, the solid angles, and the recording-channel efficiencies are maximal.

The degree of overlap g_{ie} of the volumes is always less than unity because of the presence of penumbral regions for the collimators, and amounts to 0.3 to ~ 1 in the range $\vartheta = 2$–$18°$. The solid angle of the ion collimator ($\Delta\Omega_i = 2 \cdot 10^{-4}$ sr) was chosen to satisfy the condition that the regions ($l = 4$ mm) visible by both collimators at $\vartheta = 10°$ be the same. The angular divergence of the primary beam was 4′, and that of the scattered ions, determined by the coillimator geometry, was 9′. The resolution of the experimental setup in terms of the impact parameter p was not worse than 1%.

To analyze the electrons in energy we used a high-aperture cylindrical analyzer ($\Delta\Omega_e = 0.22$ sr) with "ring-axis" entry-angle focusing similar to that used earlier to observe Auger transitions in a quasimolecule.[3] Energy resolutions of several percent are sufficient for the study of the energy spectra of quasimolecule electrons that have a continuous distribution. In experiment, the analyzer resolution $\Delta E_e / E_e$ was 4%.

The resolution time τ_r of the coincidence circuit is bounded from below by the scatter $\Delta\tau_{fl}$ of the particle flight times to the corresponding detectors and by the temporal scatter $\Delta\tau_{app}$ introduced by the recording apparatus, i.e., $\tau_r \geqslant \Delta\tau_{fl} + \Delta\tau_{app}$. The optimum resolving time ($\tau_r \simeq 60$ nsec) was determined experimentally from an analysis of the τ_r-dependence of the intensities n_{true} of the true coincidences and n_{ran} of the random ones, as well as from the minimum exposure time $t \sim n_{ran}/n_{true}^2$ needed to obtain a specified error δ.

Under conditions when n_{ran} greatly exceeds n_{true}, it becomes particularly important to decrease the errors in the measurement of the number of random coincidences. To this end we used electronic circuitry[4] in which the number of true and random coincidences was measured simultaneously and was determined by the same resolving time.

Under the chosen experimental conditions, the error δ in the measurement of the number of true coincidences amount to 10–15% within an exposure time $t = 3$–5 hr. The error in the determination of the absolute cross sections, which is connected with the geometry error and with the inaccurate measurement of the intensity of the ions of the primary beam and of the density of the gas-target atoms, amounted to $\sim 50\%$.

III. RESULTS OF INVESTIGATION OF THE QUASIMOLECULE Kr–Kr

1. Auger-electron spectra

Figure 2 shows the experimental spectra of the electrons produced in Kr^+–Kr collisions at a collision energy $E_0 = 50$ keV. The $d\sigma(E_e)/dE_e$ spectrum was taken from Ref. 3 and the $d^2\sigma(E_e)/dE_e\,dp$ spectra were measured in the

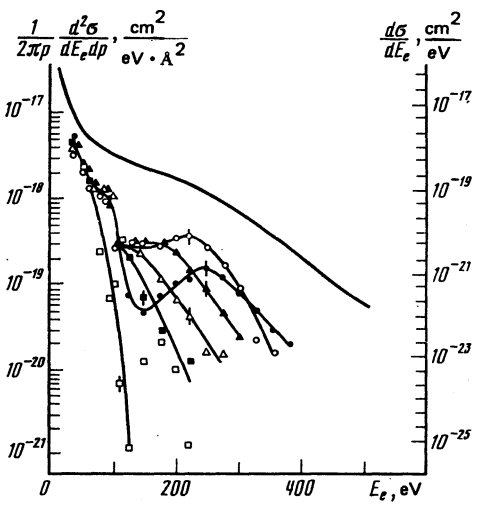

FIG. 2. Energy spectra of electrons produced in Kr^+–Kr collisions at incident-ion energies $E_0 = 50$ keV. Solid curve—$d\sigma(E_e)/dE_e$ spectrum; points—$d^2\sigma(E_e)/dE_e\,dp$ spectra measured at fixed impact parameters p in a.u.; ϑ—angle of scattering of the impinging ions in degrees: □) $\vartheta = 5$, $p = 1.0$, ■) $\vartheta = 7$, $p = 0.9$; △) $\vartheta = 9$, $p = 0.8$; ▲) $\vartheta = 12$, $p = 0.7$; ○) $\vartheta = 15$, $p = 0.6$; ●) $\vartheta = 18$, $p = 0.5$.

present study for several impact parameters p. The experimental data obtained by using the coincidence method were published in our brief communications.[5]

The broad distribution observed in the $d\sigma/dE_e$ spectrum at energies $E_e > 100$ eV is due to Auger transitions to the $4p\pi$ orbital that drops when the internuclear distance R decreases. The results obtained by using the coincidence method were convincing evidence of the quasimolecular origin of the observed electrons. In the $d^2\sigma/dE_e\,dp$ spectra one could see clearly, in place of a broad band, maxima that shifted towards higher energies E_e with decreasing impact parameter. This behavior of the electron energies is typical of the Auger decay of a vacancy on a dropping orbital.

Observation of the maximum has made it possible to determine simply and much more accurately than in Ref. 3 the energies $E(R)$ and the widths $\Gamma_A(R)$ of an autoionizing level from the experimental spectra. At a parabolic time dependence of the term energy $E(t)$ in the vicinity of the turning point R_0, the spectrum of the Auger electron from the quasimolecule is expressed in terms of an Airy function Ai(x) (Refs. 6, 7):

$$\sigma'' \equiv d^2\sigma/dE_e dp = 4p\Gamma_A n\alpha^{-2/3}\mathrm{Ai}^2\{\alpha^{-1/3}[E_e - E_e(R_0) + {}^1/_2 i\Gamma_A]\}, \qquad (2)$$

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

where n is the number of vacancies in the orbital. In the derivation of (2) it was assumed that Γ_A depends little on R. The approximation (2) has been named the simple Airy approximation.

According to (2), the spectrum of the emitted electrons has the following characteristic features. At electron energies corresponding to transitions in the vicinity of R_0, the spectrum should have a principal maximum. The argument of the Airy function vanishes at $E_e = E_e(R_0)$, since Γ_A is as a rule small and the contribution of the imaginary part to the argument can be neglected. At energies $E_e > E_e(R_0)$ (in the classically forbidden region), the Airy function tends to an asymptotic dependence and the cross section is

$$\sigma'' \sim \exp\{-{}^4/_3\alpha^{-1/2}[E_e - E_e(R_0)]^{3/2}\}.$$

The appearance of electrons with energies beyond the classical limit $E_e(R_0)$ is connected with the uncertainty principle and has been called collisional broadening.[8] In the allowed-energy region $E_e \leqslant E_e(R_0)$ the Airy function $\mathrm{Ai}^2(x)$ reaches a maximum at $x = -1.02$. At still lower E_e the cross sections expressed in terms of another asymptotic $\mathrm{Ai}^2(x)$ dependence with an oscillation phase $\sim\alpha^{-1/2}[E^e - E_e(R_0)]^{3/2}$, and the amplitude of the oscillation decreases rapidly with increasing $|E_e - E_e(R_0)|$. The onset of the oscillations is due to the interference of the waves corresponding to electrons with identical energy, emitted when the particles come close together and move apart.

2. Reconstruction of the orbital from the experimental spectrum

From the exposition in II.1 it follows that the experimental orbital $E(R)$ as a function of the internuclear distance can be reconstructed from the energies of the Auger electrons produced at the closest-approach point R_0. The energy of a level with a vacancy is equal to the energy of the Auger electron, accurate to the scatter of the binding energies of the outer electrons that participate in the transition. Since the binding energies of the outer electrons are substantially lower than the transition energy, we can assume in first-order approximation that the level energy is equal to $E_e(R_0)$. The procedure for determining $E_e(R_0)$ from the spectra $d^2\sigma(E_e)/dE_e\,dp$ is relatively simple, since it is known that the value of the function $\mathrm{Ai}^2(0)$, i.e., at the point $E_e = E_e(R_0)$, is 0.44 of the value of the function $\mathrm{Ai}^2(x)$ at the maximum.

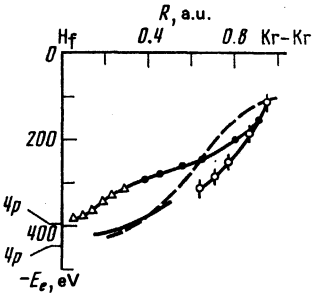

FIG. 3. Experimental level with vacancies in the Kr–Kr quasimolecules, reconstructed from the energy spectra of the electrons $d\sigma/dE_e$: ●) Ref. 3; △) Ref. 10 and $d^2\sigma/dE_e\,dp$: ○) Ref. 5; solid curve—Ref. 9; dashed—calculation.[13]

Figure 3 shows sections of the orbital obtained from the spectrum measured by the coincidence method in our studies and in the recent paper.[9] The same figure shows an orbital reconstructed from the $d\sigma/dE_e$ spectra.[3,10] One can see a satisfactory agreement between the experimental $E(R)$ dependences obtained by the two methods. At the same time it should be noted that the accuracy with which $A(R)$ is reconstructed from the spectra at fixed impact parameters is higher. In contrast to the $d\sigma/dE_e$ spectra, where the determination of $E_e(R)_0$ is difficult, the $d^2\sigma/dE_e\,dp$ spectra contain distinct maxima, from which the values of $E_e(R_0)$ can be easily obtained. At the present time, a number of calculations of the molecular orbitals were performed for the Kr–Kr system.[11–13] The orbital that agrees best with the experimental one is the one calculated in a recent paper[13] (shown in the figure).

In addition to determining the energy $E(R)$, we obtained the values of the autoionizing level width Γ_A. The values of Γ_A were obtained from the measured absolute cross sections $d^2\sigma/dE_e\,dp$ by means of Eq. (2). According to an estimate for $n = 1$, the value of Γ_A turned out to be $\sim 3\cdot 10^{15}$ sec^{-1} (~ 2 eV), i.e., the lifetime of the vacancy in the quasimolecule is comparable with the collision time. The causes of such a large width of the autoionizing level may be the increase, due to upward motion of the filled orbitals, in the number of upper-level electrons capable of participating in the Auger transitions, and the increase of the overlap of the wave functions of the states between which the transition takes place (in analogy with the Koster-Kronig transitions). An increase of Γ_A in a quasimolecule was demonstrated in a model calculation in Ref. 14.

3. Collision broadening of auger spectra

The collision-broadening effect was investigated in detail for radiative transitions. Since the cause of the collision broadening in both cases is the same—motion of the nuclei—all the regularities obtained for radiative transitions should be valid also for Auger emission. This conclusion was confirmed in Ref. 15, where collision broadening in the Auger spectra was investigated as a function of the collision velocity and of the charges of the interacting atoms. The determination of the impact parameter p in the present paper, in contrast to Ref. 15 where $d\sigma/dE_e$ spectra were measured, makes possible direct comparison of the experimental dependence of the collisional broadening on the relative velocity v_0 and of the absolute values of the broadening with the predictions of the theory. The results of the present investigations were briefly reported in Ref. 16.

In electron spectra the collision broadening manifests itself in the form of "tails" that exist beyond the maximum classically allowed energy $E_e(R_0)$ and decrease exponentially with increasing E_e. The collision broadening is customarily characterized by a quantity Γ_{CB} equal to the energy interval over which the cross section decreases to one-half compared with the value at the point $E_e(R_0)$. It follows from (2) that

$$\Gamma_{CB}=0.42\left(\frac{1}{2}\frac{dE_e}{dR}\frac{d^2R}{dt^2}\right)^{1/3}\Big|_{R=R_0}. \tag{3}$$

Since the experimental orbital for the quasimolecule Kr–Kr is well described by the relation $E(R)\sim aR^2$, and the radial acceleration is $d^2R/dt^2\sim v_0^2R_0$ in the vicinity of R_0, the collision broadening, according to (3), should be given by the relation $\Gamma_{CB}\sim v_0^{2/3}$. Such a relation was predicted for radiative transitions in Refs. 17 and 18.

Figure 4 shows the values of $\Gamma_{CB}(v_0)$ from the $d\sigma/dE_e$ spectra,[15] and also the Γ_{CB} obtained from spectra measured by the coincidence method at energies $E_0 = 25$, 50, and 100 keV at a fixed impact parameter $p = 0.75$ a.u. It can be seen that the values of Γ_{CB} obtained in the present paper lie on the theoretically predicted plot of $\Gamma_{CB}\sim v_0^{2/3}$.

At the same time, as can be seen from Fig. 4, there is a substantial difference between the values of Γ_{CB} determined in experiment and calculated by formula (3). The experimental values of Γ_{CB} turn out to be approximately three times larger than the calculated ones. The possible cause of the discrepancy will be discussed below.

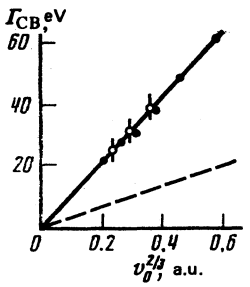

FIG. 4. Dependence of the collisional broadening Γ_{CB} of the spectra of the electrons from the quasimolecule Kr–Kr on the collision velocity v_0: ●) from Ref. 15, ○) present data, dashes—calculation by formula (3).

IV. SPECTROSCOPY OF LIGHT QUASIMOLECULES

1. Emission of electrons from light quasimolecules

The experimental method developed by us was used also to study light quasimolecules. In contrast to the quasimolecule Kr–Kr, in light quasimolecules (for example, Ne–Ne) the structure of the orbitals is much simpler, so that it is possible to identify unambiguously the levels with vacancies. In addition, for a small number of light quasimolecules there have been performed at present laborious but sufficiently straightforward calculations of the molecular orbitals, making it possible to compare directly the results of the theory and experiment. The results of investigation of the spectroscopy of light quasimolecules were published in brief communications.[19,20]

It is known that the energy spectra $d\sigma(E_e)/dE_e$ of electrons produced in collisions of light atoms ($Z\leqslant 10$) at keV initial energies consist in the region $E_e \gtrsim 30$ eV of two characteristic parts: a discrete peak connected with the Auger decay of the K vacancy after the collisions, and a considerable continuous component that decreases with increasing electron energy approximately in accord with an exponential law.

Several attempts were made to explain the nature of these continuous components. The most interesting mechanism was proposed recently in Ref. 21. Direct electron transitions into the continuum, induced by nuclear motion, from the bound states of the quasimolecule were considered. It was shown that the $d\sigma/dE_e$ spectra can be described by transitions of electrons from orbitals that move up abruptly from under the limit of the continuous spectrum when the internuclear distance is decreased.

There exists, however, also another possibility of appearance of the electrons with continuous distribution, connected with the decay of the internal vacancies in the quasimolecule via Auger transitions. In particular, in collisions of light atoms having $Z\leqslant 10$ the vacancies are produced because of the approach of the $2p\pi$ orbital, since this orbital is made up of unfilled $2p$ levels of the atoms.

2. Auger transitions to the $2p\pi$ orbital

To illustrate the possibility of formation of a continuous component on account of lowering of the unfilled orbitals, investigations were made of O^+–O_2 and Ne^+–Ne collisions in the collision-energy range $E_0 = 5$–15 keV and at impact parameters $p = 0.2$–1.4 a.u., using the coincidence method. Typical spectra produced in Ne^+–Ne collisions with impact parameter $p = 0.37$ a.u. are shown in Fig. 5. The advantages of using the cited cross sections $v_0^{4/3}p^{-1}d^2\sigma/dE_e dp$ will be explained below. It can be seen that with increasing electron energy E_e the cross section in spectrum 1 decreases approximately exponentially. At the same time, in spectra 2 and 3 maxima at $E_e \approx 80$ eV are superimposed on the exponential component. The maxima were observed in the spectra also in O^+–O_2 collisions.

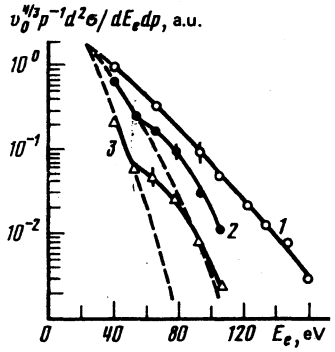

FIG. 5. Energy spectra of electrons produced in Ne^+–Ni collisions with fixed impact parameter $p = 0.37$ a.u. at different E_0 (in keV) and ϑ (in degrees): 1) $E_0 = 50$, $\vartheta = 4$; 2) $E_0 = 30$, $\vartheta = 6.7$; 3) $E_0 = 15$, $\vartheta = 13.3$.

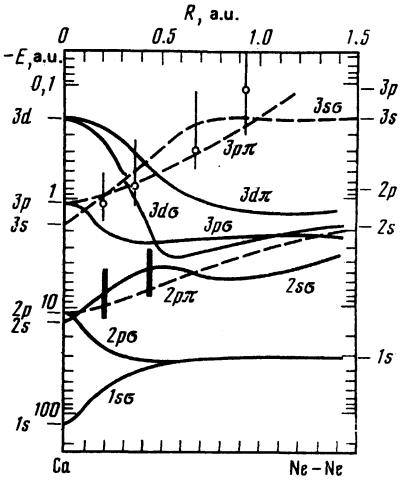

FIG. 6. MO diagram for the Ne–Ne quasimolecule.

The electron energies at the maxima differ substantially from the energies of the Auger electrons produced when the K-vacancies of isolated O atoms ($\sim$470 eV) and Ne atoms ($\sim$750 eV) are filled; on the contrary, they correspond to the energies of the Auger electrons due to the decay of $2p\pi$ vacancies, which can be expected on the basis of the correlation diagrams for the Ne–Ne quasimolecule.[22] The displacement of the maxima towards larger E_e with decreasing impact parameter has shown that the Auger transitions are produced on the dropping orbital.

To confirm the quasimolecular origin of the observed maxima we investigated the collisions Ne^+–Ne and Ne^{2+}–Ne collisions at impinging -ion energy $E_0 = 30$ keV and at $p = 0.37$ a.u. Obviously, the probability of an Auger transition in a quasimolecule is proportional to the number of vacancies on the $2p\pi$ orbital, which is determined by the number of combinations of the distribution of the $2p$ vacancies that existed in the atoms prior to the collision, over the orbitals formed out of the $2p$ levels. The statistical mean number of vacancies on a $2p\pi$ orbital in collisions with participation of the ions Ne and Ne^{2+} differ by a factor of two. The experimentally measured ratio of the cross sections for the yield of quasimolecular electrons is 1.8 ± 0.5 and agrees, within the limits of experimental error, with the predicted value.

Figure 6 shows a diagram of the quasimolecular orbitals of the system Ne–Ne. The vertical lines on Fig. 6 show the energies $E_{2p\pi}$ obtained by us from experimental data on the electron energies $E_e(R_0)$. The heights of the vertical lines correspond to the uncertainties of the binding energies of the outer electrons. As can be seen from Fig. 6, the experimental and calculated energies $E_{2p\pi}$ are in good agreement. Good agreement is likewise observed between the experimental and calculated $2p\pi$ orbital for the O–O quasimolecule.[19]

The values of Γ_A per vacancy were determined from Eq. (2). The average number of vacancies on a $2p\pi$ orbital in the investigated cases O^+–O^2 and Ne^+–Ne are respectively 1.67 and 0.33. According to an estimate, Γ_A is $\sim 10^{14}$ sec^{-1} ($\sim$0.1 eV) for the O–O quasimolecule and $\sim 10^{15}$ sec^{-1} ($\sim$1 eV) for Ne–Ne. Both values are close to the probability of Auger decay of $2p$ vacancies in the unified atoms S ($Z = 16$) and Ca ($Z = 20$). The theoretical values of Γ_A for the S and Ca atoms with $2p$ vacancies are respectively $1.0 \cdot 10^{14}$ and $3.3 \cdot 10^{14}$ sec^{-1}.

It is seen thus that the conditions that lead to an increase of the width of the autoionizing state of a quasimolecule, compared with the isolated atom, do not always exist. Indeed, in the Ca (or S) atoms produced via collision (see Fig. 6), and in an isolated atom, the number of external electrons capable of participating in the Auger decay of a $2p$ vacancy is approximately the same. The only difference is that in the quasimolecule these electrons can be on excited levels.

3. Spectroscopy of $3p\pi$ orbital of the Ne^+–Ne quasimolecule

It can be assumed that the exponential components in the spectra of the electronic $d^2\sigma/dE_e dp$ (see Fig. 5) are due to collisional broadening of the energy distribution of the electrons produced in Auger decay of vacancies on shallow (lying higher than $2p\pi$) orbitals. In contrast to Auger transitions to $2p\pi$ vacancies, in the latter case the maxima of the Airy function could not be investigated experimentally, since they are located in the region of low electron energies in which autoionization peaks connected with excitation of outer shells of the atoms predominate. A different method was proposed for determining $E_e(R_0)$.

The method is particularly simple if the straight-line-flight approximation is valid and the decrease of the number of vacancies n as a result of decay is small. In this case (2) is expressed as follows:

$$\sigma'' = 4p\Gamma_A n \left(\frac{1}{2}\frac{dE_e}{dR}\frac{v_0^2}{R_0}\right)^{-2/3} \times \mathrm{Ai}^2\left\{\left(\frac{1}{2}\frac{dE_e}{dR}\frac{v_0^2}{R_0}\right)^{-1/3}[E_e - E_e(R_0)]\right\}. \quad (4)$$

It follows from (4) directly that the spectra $v_0^{4/3}\sigma''(E_e)$, measured at one and the same impact parameter but at different velocities, intersect at the point $E_e = E_e(R_0)$, where the argument of Ai(x) vanishes. Thus, a determination of the intersection points makes it possible to obtain the $E_e(R)$ depen-

dence. The intersection points turned out to be located outside the region of the measured values of E_e and were determined by extrapolating the cross sections towards lower energies. An example of finding one value of $E_e(R)$ is shown in Fig. 5. The experimental values of $E_e(R)$ turned out to be positive in the entire interval of distances between the nuclei, i.e, the level is auto-ionizing.

The experimental level designated by circles in compared in Fig. 6 with the diagram of the molecular orbitals. It can be seen that the observed level duplicates, within the limit of errors, the course of the $3p\pi$ and $3s\pi$ orbitals that go lower with decreasing R and go over at $R = 0$ into the $3p$ and $3s$ levels of the unified atom Ca. Both orbitals are formed out of unfilled $3p$ and $3s$ levels of the Ne atoms and have four and two vacancies, respectively.

The width of the autoionizing level $\Gamma_A n$, determined from (4), is $(1.2 \pm 0.4)\cdot 10^{16}$ sec^{-1} and is independent of R within the limits of errors. If it is recognized that the total number of vacancies n on the $3p\pi$ and $3s\pi$ orbitals is six, then $\Gamma_A \approx 2\cdot 10^{15}$ sec^{-1} (~ 2 eV). The theoretical value of Γ_A for a $3p$ vacancy in the Ca ion is 0.1 eV. In the Ne–Ne quasimolecule, however, owing to the upward motion of the $4f\sigma$, $3d\pi$, and $3d\sigma$ orbitals, the number of electrons capable of participating in Auger transitions turns out to be substantially larger than for the Ca ion with a single inner $3p$ vacancy, and this may be the cause of the observed increase of Γ_A.

Thus, an investigation of light quasimolecules has shown that Auger transitions in a quasimolecule have appreciable cross sections and turn out to be a rather prevalent phenomenon. This is due primarily to the fact that a quasimolecule always has orbitals that are made up of vacant outer levels of colliding atoms and that drop lower when the internuclear distances are comparable with the dimensions of the electron shells. It can be assumed that the Auger decay of the vacancies in the quasimolecule is one of the main mechanisms of emission of electrons having a continuous energy distribution in atomic collisions.

V. COMPARISON OF EXPERIMENTAL AND THEORETICAL ELECTRON SPECTRA

The values of the energy E and of the width Γ_A of the autoionizing level of a quasimolecule as a function of the distance between nuclei were obtained under the assumption that the real spectrum is described by expression (2), obtained in the framework of the simple Airy approximation. Thus does not mean, however, that the spectrum calculated for such an "experimental" level and the measured spectrum automatically coincide. As noted above, for the Kr–Kr quasimolecule the calculated and measured spectra in the classically forbidden region $E_e > E_e(R_0)$ are noticeably different.

Figure 7 shows the calculated and experimental spectra $d^2\sigma/dE_e dp$ for O^+–O_2 and Kr^+–Kr collisions at $E_0 = 50$ keV and for the respective parameters $p = 0.3$ and 0.6 a.u. The solid line in Fig. 7a described the spectrum of the Auger electrons from the decay of the $2p\pi$ vacancy in the O–O quasimolecule, calculated within the framework of the simple Airy approximation (2) at the experimental values $\Gamma_A = 10^{-2}$ a.u. and $dE/dR_{R=34\ \text{a.u.}} = 5.2$ a.u. The calculated spectrum is superimposed on the continuous component shown by the dashed line. It can be seen that within the limits of errors the calculated spectrum agrees with the experimental one. A similar calculation is shown in Fig. 7b for the Kr–Kr quasimolecule at the experimental values $\Gamma_A = 5.2\cdot 10^{-3}$ a.u. and $dE/dR_{R=0.7\ \text{a.u.}} = 16.2$ a.u. It can be seen that the simple Airy approximation accounts sufficiently well for the general behavior of the spectrum as a function of E_e. The difference manifests itself in more subtle details—in the oscillations of the cross section at $E_e < E_e(R_0)$ and in the values of the cross section in the classically forbidden region at high energies.

It was not the task of the present paper to observe the oscillatory structure in the spectrum of the Kr–Kr quasimolecule. The point is that the measured spectrum (in contrast to the theoretical one) is broad not only on account of the instrumental function of the electron analyzer (~ 0.5 a.u.) and the Doppler effect (~ 0.3 a.u.), but also on account of the distribution of the states of the outer electrons that participate in the Auger transitions (~ 1 a.u.). These factors lead to a smearing of the oscillatory structure in our case. However, when the period of the oscillations increases, for example when the velocity of the collisions v_0 increases, such oscillations can appear. Indeed, in Ref. 9, where the spectra of the Kr–Kr quasimolecule were investigated at a collision energy 400 keV, an oscillatory picture was observed.

We have investigated the influence of the two corrections to a simple Airy approximation on the behavior of the spectrum in the classically forbidden region. The approxi-

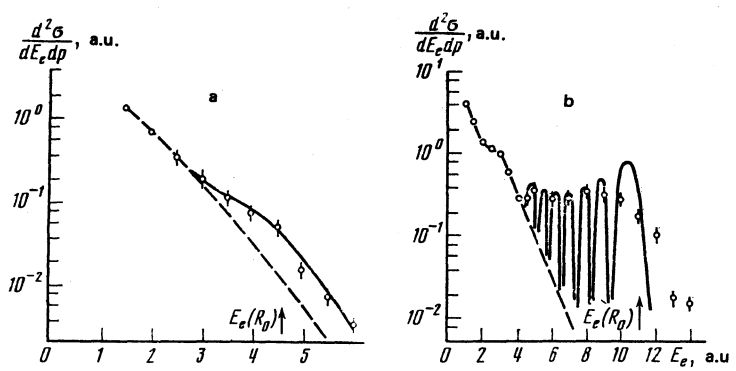

FIG. 7. Energy spectra of electrons produced in the following collisions: a) O^+–O_2, $E_0 = 50$ keV, $p = 0.3$ a.u.; b) Kr^+–Kr, $E_0 = 50$ keV, $p = 0.6$ a.u. Points—experiment, solid curve—calculation by relation (2).

mation[18] that takes into account the change of the total width of the autoionizing state (for example, as a result of a change in the number of vacancies on the quasimolecular orbital) contains the first derivative of the Airy function in the expression for the cross section (2). The correction proposed by V. N. Ostrovskiĭ and E. A. Solov'ev takes into account the deviation of the term $E(t)$ from a parabolic dependence near the boundary of the continuous spectrum and leads to the appearance of a factor that depends on E_e in front of the Airy function in expression (2). Calculations have shown that neither corrections exerts a noticeable influence on the behavior of the spectrum in the forbidden region. Better agreement with experiment can be obtained apparently within the framework of the so-called uniform approximation,[17] which takes into account the decay of the vacancies along the entire trajectory, and not only in the vicinity of the turning point. However, the use of this approximation calls for rather laborious calculations.

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