Vacancy formation in the *K* shell in collisions between heavy atoms

E. L. Duman and B. M. Smirnov

I. V. Kurchatov Atomic Energy Institute
(Submitted January 24, 1975)
Zh. Éksp. Teor. Fiz. **69**, 59–66 (July 1975)

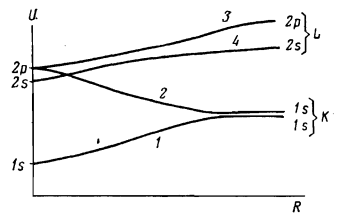
The Fano–Lichten model is used to calculate the cross section for formation of a vacancy in the *K* shell in the collision of heavy atoms. Here we consider transitions from the $2s\sigma$ or $2p\pi$ states of the quasimolecule, which correlate as $R\to\infty$ with the *L* shells of one of the partners, to the $1s\sigma$ and $2p\sigma$ states, which for an infinite separation of the nuclei go over to the *K* shell of the atom or ion. There are three mechanisms which lead to a transition between these states. Each of these mechanisms gives a substantial contribution to the cross section for vacancy formation in the *K* shell in various regions of the kinetic energy of the collision. It is shown that for a collision energy of 10–100 keV the main contribution to the cross section is from transitions associated with rotation of the axis of the quasimolecule. For the different mechanisms, analytical expressions are obtained for the cross sections for transitions leading to formation of *K* vacancies. The theoretical calculations are compared with experimental data.

PACS numbers: 34.50.H

1. In recent years a large amount of experimental data has been accumulated on ionization,[1-6] the electron spectrum,[7-14] and the x-ray spectrum[15-19] in close collisions of heavy atoms or ions with energies in the range from tens of keV to tens of MeV. On the basis of these experiments it is possible to form an idea of the physical nature of the transitions which occur on intersection of the electron shells of atomic particles. The simplest and most convenient model describing these transitions is the Fano-Lichten model.[20] According to this model the system of colliding particles is considered as a quasimolecule in which each electron moves in the self-consistent field of the core. Electron transitions between the states corresponding to this self-consistent field correspond in the final analysis to formation of vacancies in the inner shells of the colliding particles. In the present article we calculate the cross section for vacancy production in the K shell in terms of this model.

2. In the figure we have shown a diagram of the lower single-electron terms for small distances between the atoms. The numbers 1, 2, 3, and 4 in the figure for the states shown will be used later in the article. These terms correspond to the inner electrons. In the region of distances between the nuclei where these electrons interact strongly with the two nuclei, they are almost unscreened by the remaining electrons. Therefore in the first approximation we can assume that these electrons move in the Coulomb field of the nuclei. As the nuclei move together the terms approach the levels of the hydrogen-like ion with nuclear charge $Z = Z_1 + Z_2$, and for infinite separation of the nuclei the energy of the inner-electron terms approaches the energies corresponding to the states of the L and K shells of one of the partners. Thus, for $Z_1 \neq Z_2$ the $2p\pi$ and $2s\sigma$ terms are correlated as $R \to \infty$ with the L shell of the atom with charge $Z_2 > Z_1$, and the $2p\sigma$ and $1s\sigma$ terms are correlated with the K shells of the atom with charge Z_1 and the atom with charge Z_2, respectively.

Thus, remaining within the framework of the quasimolecular model, we can suggest the following mechanism for vacancy formation in the K shell for collision velocities small in comparison with the electron velocity in the K shell. On close approach of the colliding particles when the L shells of the particles intersect, a vacancy is formed in the L shell of one of the partners.



Energy of inner electrons as a function of distance between nuclei: Curve 1 corresponds to the term $1s\sigma$, curve 2 to the term $2p\sigma$, 3 to the term $2p\pi$, and 4 to the term $2s\sigma$.

On further approach this hole, which corresponds to one of the upper states in the figure, is converted as the result of transitions to a hole corresponding to one of the lower states of the figure. In the process of separating the nuclei the hole formed by this means is converted to a hole in the K shell of one of the atoms. With an approach of this type it is possible to calculate the cross section for transitions with accuracy to some factor which represents the probability W_L for formation of a vacancy in the L shell in close collisions. For atoms or ions in which the L shells are external (for example, elements of the second group in the periodic table), the cross section for K excitation can be calculated accurately in terms of this model, since an ion which already has a free L shell is taking part in the collision. In this case the probability of vacancy formation in one of the upper terms shown in the figure is determined by the statistical weight of this state.

3. For collision velocities small in comparison with the velocity of the K electron, transitions between the states considered occur at distances between the nuclei small in comparison with the dimensions of the K shell. Therefore it is convenient to expand the wave function of the colliding-particle system Ψ in wave functions φ_j of the combined atom (i.e., the atom formed on combination of the two nuclei):

$$\Psi=\sum_j C_j\varphi_j \exp\{-iE_jt\}, \tag{1}$$

where E_j is the energy of a given level on combining of the nuclei. Here and everywhere below we use the atomic system of units ($e^2 = \hbar = m = 1$).

In the region of small distances between the nuclei the Hamiltonian of the electron can be represented on the basis of perturbation theory in the form

$$\hat{H}=\hat{H}_0+\hat{V}, \tag{2}$$

where $\hat{H}_0$ is the Hamiltonian of the combined atom, and the perturbation operator V includes the variation of the Hamiltonian on separation of the nuclei.

Substituting the expansion (1) into the Schrödinger equation

$$i\hbar\partial\Psi/\partial t=\hat{H}\Psi,$$

we obtain the following system of equations for the coefficients of the expansion:

$$i\dot{C}_j=\sum_k V_{jk}\exp\{i\omega_{jk}t\}C_k-i\dot{\theta}\sum_k T_{jk}C_k\exp(i\omega_{jk}t), \tag{3}$$

where V_{jk} are the matrix elements taken from the operator of the perturbation in the wave functions of the combined atom, $\omega_{jk}=(E_j-E_k)/\hbar$, $\dot{\theta}$ is the angular velocity of rotation of the axis connecting the nuclei, and T_{jk} is the rotation matrix, which is determined by the angular eigenfunctions of the combined atom.

Using the system (3) for the states indicated in the figure, we obtain a system of five equations for the probability amplitudes C_j. One of these equations can be separated from the others in view of the symmetry of the problem, since the parity of the wave function for reflection of an electron relative to the plane of the motion is preserved. We will form two combinations of wave functions for the $2p\pi$ state, such that in the first case the fraction of the wave function dependent on the azimuthal angle φ is proportional to sin φ, and in the second case it is proportional to cos φ (the azimuthal angle is taken in the plane perpendicular to the axis joining the nuclei). Reflection of an electron relative to the plane of motion corresponds to the operation $\varphi\to-\varphi$, so that the first of the wave functions turns out to be odd relative to the reflection discussed, and the second—even. The wave functions describing the states $1s\sigma$, $2p\sigma$, and $2s\sigma$ are also even wave functions. In view of the conservation of parity in the nuclear motion, there are no transitions between the first of these states, which are $2p\pi$ combination states, and the remaining states. Consequently, there is a system of four equations which describes the transitions between the states represented in the figure.

The matrix elements of the perturbation operator V which enter into the system of equations (3) and for which general expressions have been given previously [21] can be written in the form of an expansion in powers of R as follows:

$$V_{jk}=\sum_{l=2}^{l_1+l_2}A_lZ_1Z_2\left(\frac{R}{Z}\right)^l\left(\frac{1}{r^{l+1}}\right)_{jk}[Z_2^{l-1}+(-1)^lZ_1^{l-1}] \tag{4}$$

$$-Z_1Z_2\sum_{l=1}^{l_1+l_2}A_l\frac{(2l+1)}{(l_1+l_2+n+3+l)(l_1+l_2+n+2-l)}\sum_{n=0}^{\infty}\left(\frac{R}{Z}\right)^{l_1+l_2+n+2}\cdot\Phi_n[Z_2^{l_1+l_2+1+n}+(-1)^lZ_1^{l_1+l_2+1+n}]-$$

$$-Z_1Z_2\sum_{n=0}A_0\left(\frac{R}{Z}\right)^{l_1+l_2+2+n}\Phi_n\frac{(Z_1^{l_1+l_2+1+n}+Z_2^{l_1+l_2+n+1})}{(l_1+l_2+n+2)(l_1+l_2+n+3)}.$$

Here the numbers of the states are given in the order of their increasing energy, and j and k denote the quantum numbers of the combined atom of the initial (n_1l_1m) and final (n_2l_2m) states, respectively,

$$\Phi_n=\frac{1}{n!}\frac{d^n}{dr^n}\left(\frac{\varphi_j\varphi_k}{r^{l_1+l_2}}\right)\Big|_{r=0},$$

φ_j and φ_k are the wave functions of the combined atom,

$$A_l=\frac{[(2l_1+1)(2l_2+1)]^{1/2}}{2l+1}\begin{bmatrix}l_1 & l_2 & l\\ m & -m & 0\end{bmatrix}\begin{bmatrix}l_1 & l_2 & l\\ 0 & 0 & 0\end{bmatrix}$$

[...] are Clebsch-Gordan coefficients. In finding the matrix elements it was assumed that the electron which makes a transition moves in the field of two Coulomb centers with charges Z_1 and Z_2. Therefore the matrix presented can be used wherever the splitting between the 2s and 2p levels of the combined atom can be neglected, i.e., it has the form indicated for distances between nuclei

$$(\Delta\varepsilon)^{1/2}/Z^2\ll R\ll Z^{-1}, \tag{5}$$

where $\Delta\epsilon$ is the energy difference for the 2s and 2p levels of the electron of the combined atom.

The matrix elements T^l_{ik} can be expressed in terms of the $\hat{D}^l$-rotation matrix [22]:

$$T^l_{nk}=\sum_m D^{*l}_{nm}\left(0,\frac{\pi}{2},0\right)mD^l_{km}\left(0,\frac{\pi}{2},0\right). \tag{5a}$$

Here the upper index l is the orbital angular momentum of the combined atom and the lower indices are the projections of the angular momentum l. In our case the rotation of the axis of the quasimolecule affects only two states: $2p\pi$ and $2p\sigma$, and here $T^1_{\pi\sigma}=1$, $T^1_{\sigma\pi}=-1$; the remaining matrix elements for this matrix are zero.

We are interested in the processes of transition of the hole from the state of the L shell. Accordingly in what follows we will consider only transitions from the states $2s\sigma$ and $2p\pi$ to the states $1s\sigma$ and $2p\sigma$. There are three mechanisms which lead to transitions between these states, which follow from the system of equations (3). The first of these involves rotation of the axis of the quasimolecule and leads to a transition between the states $2p\pi$ and $2p\sigma$. The role of this mechanism in formation of a hole in the K shell of the colliding particles has been discussed previously in several articles. [23,24,25] Transitions between these states as a result of rotation of the axis joining the nuclei occur at a distance R between the nuclei and with collision impact parameters ρ determined from the relation $|V_{22}(\rho)-V_{33}(\rho)|\sim\dot{\theta}=v/\rho$, i.e., $\rho\sim Z^{-1}(v/Z)^{1/2}$. Here the cross section for the process considered turned out to be of the order

$$\sigma_{32}\sim Z^{-2}(v/Z)^{2/3}. \tag{6}$$

The other mechanism involves a transition between the states $2s\sigma$ and $2p\sigma$ under the influence of the perturbation. For collision impact parameters exceeding a value $\rho\gg v/|V_{44}(\rho)-V_{22}(\rho)|$, the transitions become adiabatically improbable, i.e., the main contribution to the transition considered is from collision impact parameters

$$\rho\sim Z^{-1}(v/Z)^{1/2}.$$

The third mechanism corresponds to a transition between the states $2s\sigma$ and $1s\sigma$ and involves violation of the adiabatic approximation at small collision impact parameters $\rho\le v/\omega$, where $\omega=(3/8)Z^2$ is the energy difference of these states. [21]

4. We will find the cross sections which correspond to the different transition mechanisms between the electronic states of the quasimolecule.

Before going directly to the calculations, let us introduce parameters α and Z such that

$$Z_1-Z_2=\alpha Z,\quad Z=Z_1+Z_2. \tag{7}$$

Note that in the region of distances between nuclei which give the main contribution to the cross sections of the transitions considered, the matrix elements $V_{jk}(R)$ which determine the frequency of transitions between the states of the quasimolecule are much less than unity and less than the frequency of rotation of the axis joining the nuclei:

$$V_{jk}\ll 1, \quad \theta > V_{jk}.$$

This fact allows us to separate from the system of equations (3) the equations which describe the rotation of the quasimolecule axis. For this reason the transition amplitudes not associated with rotation of the quasimolecule axis can be found on the basis of perturbation theory. Using perturbation theory, we obtain for the amplitudes of transitions to state (4):

$$C_{42}\cong -i\int V_{42}\exp\left\{-i\int_{-\infty}^{t}\frac{V_{44}-V_{22}}{2}dt'\right\}dt\cong -i\frac{3\sqrt{\pi}}{4}\alpha\left[\frac{120}{(1-\alpha^2)}\frac{v}{Z}\right]^{1/3}x^{3/2}\Phi(x)$$

$$=-i\frac{\sqrt{3}}{4}\alpha\left[\frac{120}{(1-\alpha^2)}\frac{v}{Z}\right]^{1/3}x^2K_{1/3}\left(\frac{2}{3}x^{3/2}\right), \qquad (8)$$

$$C_{41}\cong -i\int V_{41}e^{-i\omega t}\,dt=-i\frac{\sqrt{2}}{8}\frac{\rho^2(1-\alpha^4)vZ^5}{\omega^2}K_2\left(\frac{\rho\omega}{v}\right). \qquad (9)$$

Here

$$x=[(1-\alpha^2)Z^4/120v]^{2/3}\rho^2,$$

ρ is the impact parameter, $K_{1/3}(x)$ is the MacDonald function. As a result of these transitions the hole in the L shell of one of the atoms goes over to a hole in the K shell. Note that for identical charges of the nuclei $Z_1 = Z_2$ the matrix element of the transition vanishes, $V_{42} = 0$, i.e., transitions of this type do not occur.

For the amplitudes of transitions between states 2 and 3 as the result of rotation of the quasimolecule axis, we have the following system of equations:

$$iC_2=-BR^2C_2+i\frac{\rho v}{R^2}C_3, \quad iC_3=BR^2C_3-i\frac{\rho v}{R^2}C_2, \quad B=\frac{Z^4(1-\alpha^2)}{160}. \qquad (10)$$

The law of motion of the nuclei in a Coulomb field has the form [21]

$$R=\rho[(1+\beta^2)^{1/2}\operatorname{ch}\xi+\beta], \quad t=\frac{\rho}{v}[(1+\beta^2)^{1/2}\operatorname{sh}\xi+\beta\xi], \qquad (11)$$

where ξ is a dimensionless variable, $-\infty \le \xi \le \infty$, $\beta = a/\rho$, $a = Z_1Z_2/2E$, E is the kinetic energy of the collision.

Solution of the system of equations (10) can be carried out by a method similar to that of Vaĭnshteĭn, Presnyakov, and Sobel'man. [26] This gives the following approximate value for the transition probability amplitude:

$$C_{32}\approx \sin\int_{-\infty}^{\infty}\frac{\rho v}{R^2}\exp\left\{4iB\int_{-\infty}^{t}R^2\,dt'\right\}dt. \qquad (12)$$

Substituting into Eq. (12) the law of motion (11), we find the transition probability amplitude:

$$C_{32}\approx \sin\frac{\pi}{(1+3a^2/4\rho^2)^{1/2}}\exp\left[-\frac{8}{3}\frac{B\rho^3}{v}\left(1+\frac{a\pi}{2\rho}\right)\right]. \qquad (13)$$

Equation (13) for the transition amplitude takes into account curvature of the trajectory. In the limiting case of high energies when the trajectory curvature can be neglected, this formula takes the form

$$C_{32}=C_{32}^{(0)}=\sin[\pi\exp(-{}^8/_3B\rho^3/v)]. \qquad (14)$$

Substituting the values of the transition probability amplitudes (8), (9), and (13) into the expression for the transition cross sections

$$\sigma_{ik}=2\pi\int_0\rho\,d\rho|C_{ik}|^2, \qquad (15)$$

we find

$$\sigma_{41}\cong\frac{2\pi Z^{10}}{5\omega^{10}}v^8=\frac{2.3\cdot 10^4(1-\alpha^4)^2}{Z^{10}}v^8, \qquad (16)$$

$$\sigma_{42}\cong\frac{(3)^{10/3}\pi\Gamma^2(5/3)\Gamma(4/3)\alpha^2}{64\Gamma(10/3)Z^2}\left[\frac{120}{(1-\alpha^2)}\frac{v}{Z}\right]^{4/3}=\frac{1}{2}\frac{(Z_1-Z_2)^2}{Z^4}\left[\frac{120v}{Z}\right]^{4/3}, \qquad (17)$$

$$\sigma_{32}\cong\frac{\pi}{Z^2}\left[\frac{80v}{(1-\alpha^2)Z}\right]^{2/3}\left[1+\frac{3(1-\alpha^2)Z^6}{256E^2[80v/(1-\alpha^2)Z]^{2/3}}\right]^{-1}$$

$$\times\exp\left\{-\frac{1.4\pi(1-\alpha^2)Z^3}{8E[80v/(1-\alpha^2)Z]^{1/3}}\right\} \qquad (18)$$

where E is the collision energy. The cross section σ_{41} was calculated previously in the work of one of the authors. [21]

The total cross section for K excitation is determined on the basis of the cross sections found as the sum of the partial cross sections (16)—(18) with a weight equal to the probability W_L for vacancy formation in the L shell. This state of the L shell is one of the upper states in the figure. Accordingly, the cross section for vacancy formation in the K shell of one of the atoms can be written in the form

$$\sigma_K=\sum_L W_L\sigma_{KL}, \qquad (19)$$

where σ_{KL} is the cross section for the transition between corresponding states of the figure, one of which corresponds to the K shell and the other to the L shell of the colliding partners.

5. Two processes compete in filling the vacancies in the inner shells: The energy liberated in filling a vacancy can be converted into ionization of the outer electrons—Auger ionization, [7-14] and hard x rays are produced. [16-19] Therefore the process being investigated can be written in the form

$$A^{+}+B\to A^{n+}+B^{m+}+(m+n-1)e+h\nu_X. \qquad (20)$$

Measurement of the intensities of the emitted electrons permits determination of the absolute values of the Auger-process cross section σ_A, and measurement of the intensity of the x rays provides the possibility of determining the cross section for emission of an x-ray photon σ_X. The cross section for vacancy formation in the inner shell σ_I is the sum of these cross sections and is equal to

$$\sigma_I=\sigma_A+\sigma_X.$$

The customary parameter of the process considered is the x-ray quantum yield:

$$\omega=\sigma_X/\sigma_I=\sigma_X/(\sigma_X+\sigma_A).$$

The difficulty in the experiment is that measurements of σ_X and σ_A are based on different experimental techniques. Therefore none of the experimental studies mentioned simultaneously measure the values of σ_X and σ_A within a single experiment. In most of the experiments performed, one of these quantities was measured, and more frequently—its relative value.

The theory which has been developed here permits reliable determination also of the relative values for the cross sections for vacancy formation in the K shell, since only the quantity W_L—the probability of formation of a vacancy in the L shell—is left undetermined. In the simplest cases we can correctly determine these quantities. Thus, in the case of the collision Ne^+-Ne we will

E, keV	Cross section for K-vacancy formation, 10^{-19} cm^2						
	Ne+—Ne			C+—C			
	Theory, Eq. (18), $W_L\sigma_{32}$	Experiments [12,16]		Theory		Experiment [15]	
		$\sigma_I = \sigma_X + \sigma_A$	$\omega \cdot 10^2$	Equation (18)	Watanabe [25]	$\sigma_I = \sigma_X/\omega$	ω [27]
20	–	–	–	3	24	2.8	$9 \cdot 10^{-4}$
50	0.07	0.03	1.6	8	59	11	
75	0.19	0.18 *	2.4	11	55	18	
100	0.29	0.27	3.2	13	64	27	

*For an energy E = 75 keV the K-ionization cross section σ_I was found from the known experimental cross section [16] σ_X(E = 75 keV) = 4.2×10^{-22} cm^2 for a fluorescence yield value $\bar{\omega} = 2.4 \times 10^{-2}$ from the formula $\sigma_I = \sigma_X/\bar{\omega}$.

assume $W_L = (\bar{n} + 1)/16$, where $\bar{n} = 3$ is the most probable number of liberated electrons [6] on intersection of the L shells of the colliding particles, and this quantity is assumed identical for states 3 and 4. In the case C^+-C in accordance with Watanabe [25] we will assume $W_L = 1$. On the basis of Eqs. (16)—(18) we find that the most effective transitions with formation of K vacancies for collision energies of 20—100 keV are determined by 3—2 transitions. In the table we have given a comparison of theory and experiment for the vacancy-formation cross section in the K shell in this energy region.

[1] V. V. Afrosimov, Yu. S. Gordeev, V. K. Nikulin, A. M. Polyanskiĭ, and A. P. Shergin, Zh. Eksp. Teor. Fiz. **62**, 848 (1972) [Sov. Phys.-JETP **35**, 449 (1972)].
[2] V. V. Afrosimov, Yu. S. Gordeev, A. M. Polyanskiĭ, and A. P. Shergin, Zh. Eksp. Teor. Fiz. **63**, 799 (1972) [Sov. Phys.-JETP **36**, 418 (1972)].
[3] G. M. Thomson, P. C. Laudieri, and E. Everhart, Phys. Rev. **A1**, 1439 (1970).
[4] R. C. Der, T. M. Kavanagh, J. M. Khan, B. P. Curry, and R. J. Fortner, Phys. Rev. Lett. **21**, 1731 (1968).
[5] H. Tawara, K. Ishii, S. Morita, H. Kaji, C. N. Hsu, and T. Shiokawa, Phys. Rev. **A9**, 1617 (1974).
[6] B. Fastrup, G. Hermann, Q. Kessel, and A. Crone, Phys. Rev. **A9**, 2518 (1974).
[7] B. Fastrup, G. Hermann, and K. J. Smith, Phys. Rev. **A3**, 1591 (1971).
[8] F. W. Bingham, Phys. Rev. **182**, 180 (1969).
[9] M. E. Rudd, Proc. VII ICPEAC, Amsterdam, 1971, p. 107.
[10] D. J. Volz and M. E. Rudd, Ph. D. Thesis, Lincoln, Nebraska, 1968.
[11] G. N. Ogurtsov, I. P. Flaks, S. V. Avakyan, and N. V. Fedorenko, ZhETF Pis. Red. **8**, 541 (1968) [JETP Lett. **8**, 330 (1968)].
[12] R. C. Cacak, T. Jorgensen, Jr., and M. E. Rudd, Ph. D. Thesis, Lincoln, Nebraska, 1969.
[13] G. N. Ogurtsov, I. P. Flaks, and S. V. Avakyan, Zh. Tekh. Fiz. **40**, 2124 (1970) [Sov. Phys.-Tech. Phys. **15**, 1656 (1971)].
[14] Yu. S. Gordeev and G. N. Ogurtsov, Zh. Eksp. Teor. Fiz. **60**, 2051 (1971) [Sov. Phys.-JETP **33**, 1105 (1971)].
[15] R. J. Fortner, B. P. Curry, R. C. Der, T. M. Kavanagh, and J. M. Khan, Phys. Rev. **185**, 164 (1969).
[16] F. W. Saris and D. Onderdelinden, Physica **49**, 441 (1970); **52**, 290 (1971).
[17] A. D. Marwick, J. Phys. **B6**, 2445 (1973).
[18] R. J. Fortner and J. D. Garcia, J. Phys. **B6**, L346 (1973).
[19] S. Sackmann, H. O. Lutz, and J. Briggs, Phys. Rev. Lett. **32**, 805 (1974).
[20] U. Fano and W. Lichten, Phys. Rev. Lett. **14**, 627 (1965).
[21] B. M. Smirnov, Opt. Spektrosk. **17**, 504 (1964) [Opt. Spectrosc.].
[22] E. L. Duman, Opt. Spektrosk. **35**, 193 (1973) [Opt. Spectrosc.].
[23] D. Bates and D. A. Williams, Proc. Phys. Soc. **83**, 425 (1964).
[24] J. S. Briggs and J. Macek, J. Phys. **B6**, 982 (1973); **B5**, 579 (1972).
[25] T. Watanabe, M. Koike, and F. Minami, J. Phys. Soc. Jap. **34**, 781 (1973).
[26] L. Vaĭnshteĭn, L. Presnyakov, and P. Sobel'man, Zh. Eksp. Teor. Fiz. **43**, 518 (1962) [Sov. Phys.-JETP **16**, 370 (1963)].
[27] R. W. Fink, R. C. Jopson, H. Mark, and C. D. Swift, Rev. Mod. Phys. **38**, 513 (1966).

Translated by C. S. Robinson
6