Formation of vacancies in inner electron shells of fragments in fission of heavy elements

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The degree of ionization and the formation of vacancies in the inner electron shells of fragments in fission of ^{235}U nuclei are discussed on the basis of a diabatic correlation diagram of the weakly asymmetric system Sr–Xe. The combined degree of ionization of the fragments is 46, which agrees with the experimental data. In the M shell of the heavy fragment, vacancies are produced with almost unit probability. The probability of vacancy formation in other shells is discussed.

1. Recently there has been an extensive investigation of processes involving multiply charged ions. Special attention has been devoted to collisions of atoms in which the nuclei approach within distances less than the radius of the K shell. Of the new interesting effects in this area we note first of all the formation of vacancies in the inner electron shells[1] and the production of positrons in collisions of sufficiently heavy ions.[2] The electron and photon quasimolecular spectra which are produced in such collisions can serve as a unique source of information on the properties of the nuclei of superheavy elements.[3]

The theoretical study of such processes is greatly simplified as the result of the possibility of using the adiabatic approximation for description of the behavior of the inner-shell electrons. In this connection it is of interest to consider, from the point of view of the adiabatic approximation and of the methods which can be used in the theory of collisions of multiply charged ions, the behavior of the electron shell in fission of heavy elements such as ^{235}U, which can be discussed as "half" of a collision. In this way we have the possibility of comparing half and whole collisions at equivalent energies for the same elements and thus studying the details of the collision process, in particular the role of nonadiabatic transitions involving rotation of the internuclear axis. We note that the problem of the distribution of muons between the fission fragments in muonic atoms, which is similar in formulation, has been discussed by Demkov *et al.*,[4] also in the framework of the adiabatic approximation. Emel'yanov and Kraĭnov[5] have discussed the problem of the degree of ionization of the fragments in fission of ^{235}U in the sudden-shaking (instant-perturbation) approximation. This approach cannot be used to consider the behavior of the inner-shell electrons and, as will be evident below, is inadequate even for calculation of the degree of ionization of the fragments.

2. In fission of the nuclei of uranium atoms, multiply charged ions of the fragments with charge $q = Z - N_e$ are formed[6] (Z is the charge of the nucleus of the fragment and N_e is the total number of electrons in the ion), which fly apart with a relative velocity $v^n \approx 11$ atomic units (the velocity of the heavy fragment is $v_H^n \approx 4.5$ atomic units). For the case of propagation of the fragments in the medium a theoretical estimate of q was first given by Bohr,[7] whose discussion was based on assumption of removal of those electrons of a fragment whose orbital velocity satisfies the conditions $v_{L,H}^{el} < v_{L,H}^{n}$, respectively. In the present work we consider the state of the electron shells of the fragments directly after fission of ^{235}U, i.e., before interaction of the fragments with atoms of the medium.

We shall divide the electrons in the uranium atom into two groups: outer electrons, for which the orbital velocity is $V_U^{el} < V_H^n$ (these are electrons with $n_U 5$), and inner electrons. In accordance with the basic postulate of Ref. 7, but for the combined atom (the ^{235}U atom) we shall assume that the outer electrons, of which there are 32, are completely removed in fission of the nuclei. As was mentioned above, the starting point for calculation of the ionization cross section in Ref. 5 was the approximation of sudden perturbation for electrons with $n_U 5$. As a result of a computer calculation it was established that the charge of the light fragment is $q_L \approx 21$–22. Since the inner electrons were not considered, the calculations of Ref. 5 give a clearly underestimated value $q_H \approx 10$.

The behavior of the inner electrons, with respect to which the motion of the nuclei of the fragments can be considered to be adiabatic, we shall consider by means of a diabatic correlation diagram of the weakly isometric system Sr–Xe (the most probable fission products); see Fig. 1. With accuracy to an unimportant shift of the energy levels, this diagram coincides with that constructed by Eichler *et al.*[8] for the Kr–Xe system on the basis of a model with variable screening. As was established in the book by Nikitin and Umanskiĭ,[9] such diagrams take into account most completely all forms of interactions in the various regions of internuclear distances.

Consideration of the correlation diagram shows[1)] that in fission of ^{235}U there is a removal of electrons which fall into the diabatic terms $4s\sigma$, $4p\pi$, $4d\delta$, $4f\varphi$ which correlate with highly excited states of the fragments (for these states $v_{L,H}^{el} \ll v_{L,H}^{n}$). A similar ejection leads to removal of another 14 electrons. We note that from the point of view of the correlation diagram there is also a practically complete removal of the outer electrons with $n_U \geqslant 5$.

The value of the combined fragment charge $q_\Sigma = 46$ which follows from consideration of the correlation diagram is approximate in the respect that it does not take into ac-

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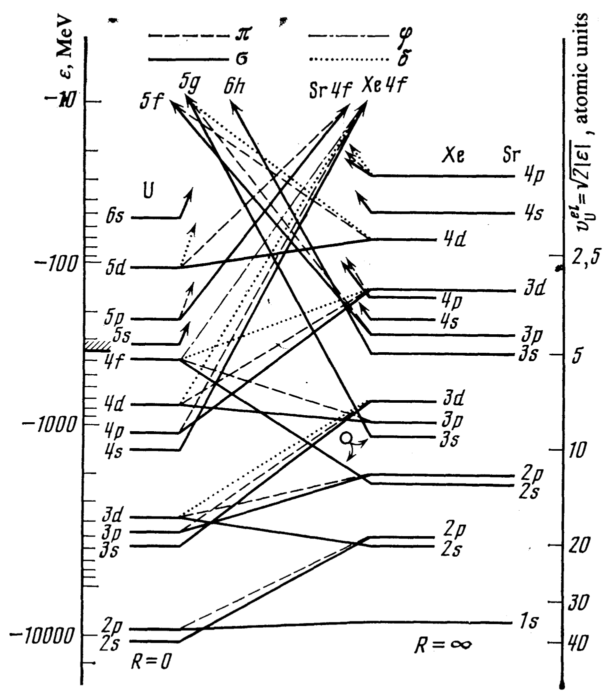

FIG. 1. Diabatic correlation diagram of the Sr–Xe system. On the energy scale we have marked the boundary between outer and inner electrons in the uranium atom. The hollow circle with two arrows illustrates the "separation" of the vacancies in the $5g\sigma$ orbital.

count either the possible stripping from upper filled orbitals with $n_U = 4$ or the capture of electrons from the orbitals $4s\sigma$, $4p\pi$, $4d\delta$ and $4f\varphi$. These processes determine the population of the states with $n_H \geqslant 4$ and $n_L \geqslant 3$, between which one can have transition of electrons according to the Demkov-Nikitin mechanism with a probability $p \approx 1$, which makes it difficult to give a final value of q for each fragment individually.

The experimental value[11] $q_\Sigma \approx 42$ was determined after passage of the fragments through the source—a layer of ^{235}U atoms with characteristic density 10^{-5} g/cm^2. The discrepancy with the value $q_\Sigma = 46$ established above can be explained by a further decrease of the fragment charge as the result of charge exchange in the atoms of the medium. This decrease is just $\Delta q_\Sigma = 4$–5 in the usual estimate of the charge-exchange cross section $\sigma \geqslant 10^{-16}$ cm^2.

3. Thus, analysis of the division of the electron shell of an atom on fission of the nucleus on the basis of a correlation diagram gives a completely reasonable estimate of q_Σ. It is particularly convenient to use a diabatic diagram in discussion of the formation of vacancies in fission of ^{235}U. As the figure shows, vacancy formation is possible in the M_H shell of the heavy fragment. A calculation of the adiabatic terms of the Kr–Xe system,[8] which is close to the one considered, permits evaluation of the strength of Landau-Zener transitions at the intersection of the $5g\sigma$ orbital with other orbitals. The greatest probability of a transition from this orbital does not exceed 0.1 (the intersection with the $4p\sigma$ orbital), which enables us to conclude that there is almost 100% probability of the formation of two vacancies in the M_H shell. In the region of formation of orbitals of the separated atoms (fragments) transition of these vacancies to the L_L shell of the light fragment is possible. An estimate on the basis of the Demkov model gives a probability $p \approx 0.5$ for formation of a vacancy in the L_L shell. Formation of vacancies in the L_L shell is possible also as the result of transition of an electron into the continuum from the $4f\sigma$ orbital in the initial stage of separation.[2)] Then with a probability $p \approx 0.1$ this vacancy, which is already in the region of formation of the orbitals of the separated atoms, will go over to the L_H shell.

Let us discuss now the possibility of experimental verification of the quasimolecular mechanism suggested above for formation of vacancies in fission of nuclei. The most systematic approach would be to measure the degree of ionization and the x radiation accompanying fission in the gas phase. Here it is necessary to take into account carefully the influence of internal conversion, which leads to radiation from the K, L, and M shells at a time 10^{-9} sec after the fission. The probability of vacancy production as the result of conversion will depend substantially on the nuclear-transition energy, the atomic number of the fragments, and the multipolarity of the γ radiation. The formation of vacancies according to the quasimolecular mechanism, on the other hand, will depend only weakly on the characteristics of the nuclei of the fragments and leads to radiation from the inner shells L and M (but not K) in 10^{-14}–10^{-13} sec.

The occurrence of L radiation of the light fragment ($E \approx 2.1$ keV) and of the heavy fragment ($E \approx 3.6$ keV) has been established experimentally,[13,14] but in these studies conversion was named as the most probable cause of the radiation. It would be interesting to attempt to observe experimentally radiation from the M shell, since in this shell in fission of ^{235}U vacancies are formed with almost unit probability, whereas vacancy formation as a consequence of conversion in the M shell is significantly less likely than in the L shell.

The quasimolecular mechanism established here for production of vacancies in the inner electron shells of atoms can occur also in other fission processes. Its characteristic feature in comparison with the well known laboratory methods of vacancy formation (collisions of heavy ions, laser sparks, and so forth) is that the energy necessary for formation of the vacancies comes directly from the nuclear process.

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[1)] Correlation diagrams are widely used for analysis of vacancy production in inner electron shells in collisions of atoms; see for example Ref. 10. In our case the analysis is greatly simplified since it is not necessary to consider transitions involving rotation of the internuclear axis.

[2)] A recent calculation[12] of the probability of direct ionization of the $4f\sigma$ orbital in head-on collisions Ag + Ag and Ag + I with $v \approx 5$ atomic units permits the conclusion that there is almost unit probability of such a transition.

[1]E. H. S. Burhop, Adv. At. Mol. Phys. **15**, 329 (1979).

[2]H. Bokemeyer *et al.*, in: Quantum Electrodynamics of Strong Fields, ed. by W. Greiner, Plenum, N. Y., 1982, p. 273.

[3]C. Stoller, W. Wölfli, G. Bonani, M. Stöckli, and M. Suter, J. Phys. B: Atom. Mol. Phys. **10**, L347 (1977).
[4]Yu. N. Demkov, D. F. Zaretskiĭ, F. F. Karpeshin, M. A. Listengarten, and V. N. Ostrovskii, Pis'ma Zh. Eksp. Teor. Fiz. **28**, 287 (1978) [JETP Lett. **28**, 263 (1978)].
[5]V. M. Emel'yanov and V. P. Kraĭnov, Yad. Fiz. **31**, 899 (1980) [Sov. J. Nucl. Phys. **31**, 464 (1980)].
[6]N. A. Perfilov, Dokl. Akad. Nauk SSSR **28**, 426 (1940).
[7]N. Bohr, Phys. Rev. **58**, 654 (1940).
[8]J. Eichler, U. Wille, B. Fastrup, and K. Taulbjerg, Phys. Rev. **A14**, 707 (1976).
[9]E. E. Nikitin and S. Ya Umanskiĭ, Neadiabaticheskie perekhody pri medlennykh atomnykh stolknoveniyakh (Nonadiabatic Transitions in Slow Atomic Collisions), Atomizdat, Moscow, 1979, p. 130.
[10]R. H. Mokler and F. Folkmann, in: Topics in Cur. Phys. **5**, 201 (1978).
[11]N. O. Lassen, K. Dan. Vidensk. Selsk. Mat.-Fys. Medd. **26**, No. 5 (1951).
[12]U. Wille, J. Phys. B: Atom. Mol. Phys. **16**, L275 (1983).
[13]R. E. Carter, J. J. Wagner, and M. E. Wyman, Bull. Amer. Phys. Soc., Ser. II, **3**, 228 (1958).
[14]R. B. Leachman, Second Intern. Conf. on Peaceful Use of Atomic Energy, Selected Reports of Foreign Scientists, Vol. 2, Neutron Physics. Atomizdat, Moscow, 1959, p. 282.

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