Energy dependence of the molecular-orbital x-ray interference structure in U^{92+}-Pb collsions

K. Schulze, J. Anton, W.-D. Sepp, and B. Fricke
Fachbereich Physik, University of Kassel, 34109 Kassel, Germany
(Received 3 November 1997)

A systematic spectroscopy of the inner shells in superheavy quasimolecule systems with energy eigenvalues in the vicinity of the negative continuum has still not been realized. The investigation of quasimolecular x rays in collisions with bare projectiles represents a unique tool to study the transiently formed molecular orbitals. For heavy-ion collision systems it seems to be a promising approach to get information about the inner orbitals at small internuclear distances. We present calculations of quasimolecular x rays from the heavy-ion collision system U^{92+}-Pb and study the dependence on the impact energy and the impact parameter. We find that up to an impact energy of 50 MeV/u the interference structure is visible in the spectrum. [S1050-2947(98)05106-3]

PACS number(s): 31.30.Jv, 31.15.Ar, 34.70.+e, 34.90.+q

The measurement of quasimolecular x rays with slow H-like or fully stripped projectiles for a defined impact parameter leads to a very distinct structure in the spectra since the photon can be emitted on the incoming and outgoing parts of the trajectory. The interference structure was measured and evaluated in the Cl^{16+}-Ar system [1] leading to the experimental determination of the $2p\pi$-$1s\sigma$ transition energy as a function of the internuclear distance [2,3]. We have calculated th x-ray emission probability for this system using a basis of many-electron two-center Dirac-Fock-Slater wave functions and including radial as well as rotational coupling [4]. Qualitatively good agreement was found with the experimental results, but depending on the impact energy, deviations occurred in the absolute intensity greater than the systematic error given by the experimentalists [3]. The reason for this deviation was the application of the one-active-electron approximation, which does not take into consideration the occupation of the orbitals due to additional electrons in the collision system.

To investigate the level behavior and electronic excitations of superheavy quasimolecules, mainly K-hole production rates, δ-electron emission, and positron creation have been studied (a comprehensive review has been given recently by Müller-Nehler and Soff [5]), but especially for highly ionized projectiles quasimolecular x rays represent a powerful tool to investigate the transiently formed molecular orbitals. In comparison to δ-electron emission and positron creation the investigation of quasimolecular x rays has the strong advantage that mainly inner orbitals, which are the subject of interest, are involved in the physical process. While in K-hole production or even K-K vacancy transfer measurements the occupation is tested only in the separated atom limit, it is tested during the whole collision by measuring the molecular-orbital (MO) x rays.

It was discussed that the interference structure could be used for the investigation of nuclear reaction times [6,7] and for testing the behavior of the $1s\sigma$ molecular level for heavy-ion collision systems at small internuclear distances [2] since it was measured by Tserruya *et al.* [1]. It has still not been realized though. On the theoretical side, Kirsch and co-workers [6] have calculated the x-ray emission probability for 5.8-MeV/u Pb-Pb collsions for varying nuclear sticking times. For the calculation of the time-dependent electronic wave function they neglected the rotational coupling [8] or assumed an impact parameter of zero [6] so that the rotational coupling vanishes. The heaviest system investigated experimentally is the system Kr^{35+}-Mo for the collision energy $E_L = 462$ MeV [7]. However, for this system the chosen impact energy is too high to find the interference structure. The minimum is shifted to energies below the characteristic lines. This shift of the interference structure to lower energies with increasing impact energies was seen in the spectra for Cl^{16+} on Ar [3] as well and reproduced by our calculation [4].

In this paper we analyze the MO x-ray spectra of the heavy-ion collision system U^{92+}-Pb and study the dependence on the impact energy and the impact parameter. We calculate the emission probability in first order with respect to the interaction with the radiation field and using the long-wavelength approximation. The main formulas of the theoretical method for the calculation of the MO spectra are summarized in [4]. The intensity of the radiation is now evaluated in the many-particle framework described in [9]. The details will be given elsewhere.

We solve the two-center Dirac-Fock-Slater equation for the molecule $(UPb)^{118+}$ with the MO-LCAO (linear combination of atomic orbitals) method and since we are essentially interested in the innermost orbitals, we restrict the basis set to the $1s_{1/2}$-$4p_{3/2}$ orbitals of U and Pb and the $1s_{1/2}$-$4d_{7/2}$ orbitals of the monopole basis set centered at the center of charge. The basis sets are adapted to the internuclear distance by taking into account the monopole parts of the potential of the collision partners into the construction of the basis functions [10]. The correlation diagram resulting from calculations at about 130 internuclear distances is shown in Fig. 1. The two-center calculation is compared with a calculation in the monopole approximation for the two-center potential. One can see that for $R > 0.02$ a.u. the deviations are no longer negligible and one has to recall that the couplings-between the orbitals have completely different forms.

We solve the coupled-channel equations with inclusion of all dynamic couplings in the subspace spanned by the mo-

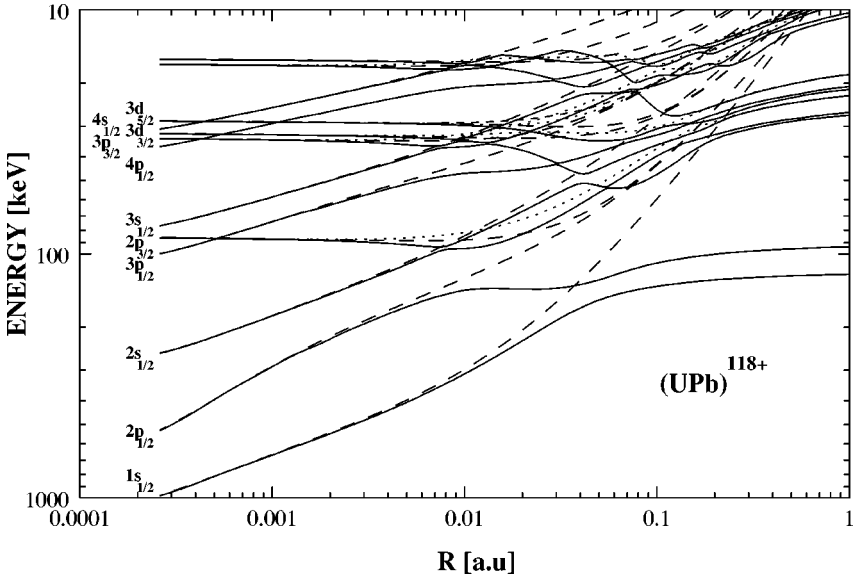

FIG. 1. Correlation diagram for the system $(\mathrm{UPb})^{118+}$ calculated with a two-center potential and in the monopole approximation. The dashed lines correspond to the monopole calculation. The two-center calculations are divided into the $m_j=\frac{1}{2}$ states (solid lines), the $m_j=\frac{3}{2}$ states (dotted lines), and the $m_j=\frac{5}{2}$ states (dashed lines). The energy eigenvalues and the corresponding molecular orbitals have been calculated by solving the two-center Dirac-Fock-Slater equation at about 130 internuclear distances. The 56 electrons taken into account are assumed to be in the ground state and the nuclei are taken as homogeneous charged spheres.

lecular orbitals corresponding to the K, L, and M shell of the united atom. In the limit $R\rightarrow\infty$ these channels correspond to the K- and L shells of U and Pb and the U $3s_{1/2}$ orbital. To get realistic results we initially occupy all channels in our basis set corresponding to the Pb target. All our calculations represent single collision processes.

We start with the analysis of the collision energy dependence of the MO x-ray spectra. Figures 2 and 3 show the emission probability as a function of the photon energy for impact energies from 3 MeV/u up to 100 MeV/u, for an impact parameter of 20 fm, and for an emission angle of $\Theta=90°$ with respect to the beam axis in the laboratory system. The spectra show a very strong dependence on the impact energy. A very distinct structure can be found only for impact energies up to 10 MeV/u; already for 20 MeV/u the strongest peak in the spectra has been shifted below photon energies that correspond to characteristic lines. For impact energies 20–50 MeV/u still a structure can be found in the spectra before the x-ray emission probability as a function of the photon energy turns into a purely exponential behavior for collision energies above 60 MeV/u.

The principal behavior of the spectra for changing impact energy can be explained by inspecting the phase difference for a two-level system. In a slow collision the phase changes

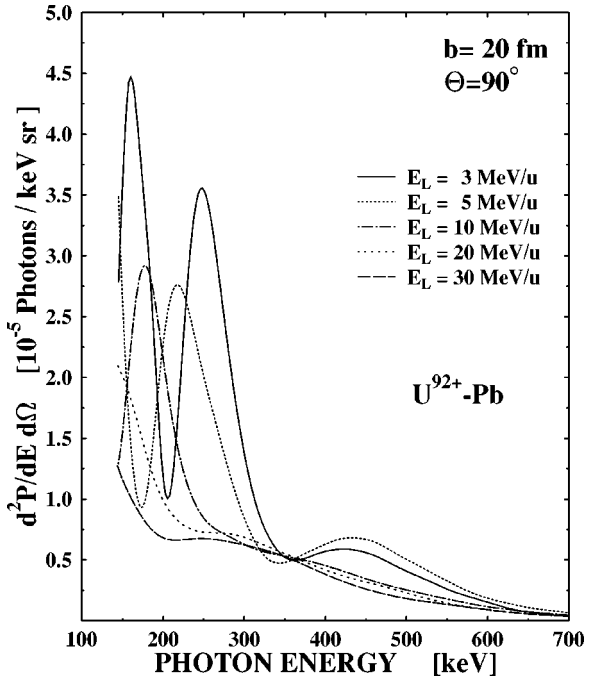

FIG. 2. Calculated double differential emission probability as a function of the photon energy for the system U^{92+} on Pb for different impact energies E_L. Twenty-eight molecular orbitals and ten electrons have been included in the calculations. The photon is emitted at an angle of $\Theta=90°$ with respect to the beam axis in the laboratory system. The collision plane is taken to be undetermined with respect to the photondetector, which results in averaging over the azimuthal angle. All calculations are done for an impact parameter of $b=20$ fm.

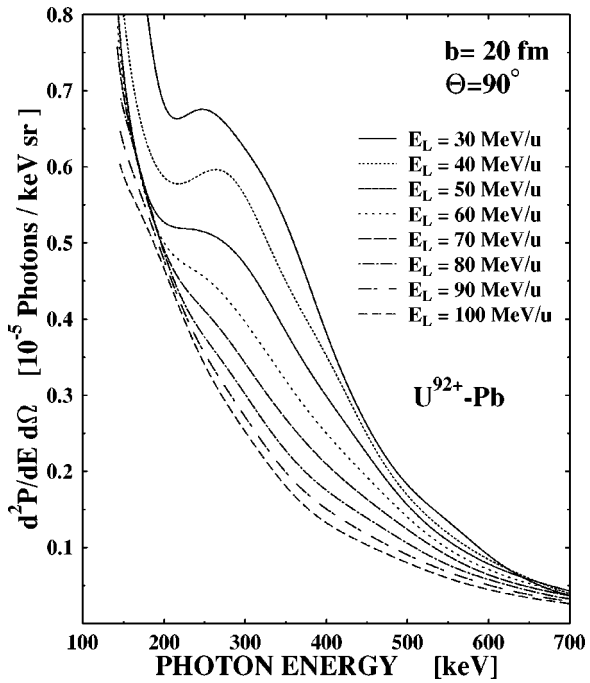

FIG. 3. Same as in Fig. 2, but for impact energies from 30 MeV/u up to 100 MeV/u. Note that the scale is changed in this figure.

more due to the lower velocity over the molecular region of the collision than in a fast collision. This results in an oscillation frequency in the spectra that decreases for increasing impact energies. This is the behavior we find. However, the striking structure of the quasimolecular spectra gets lost with increasing impact energy faster than one might expect from an experiment [11] where differential state selective cross sections for the reaction $He^{2+} + He \rightarrow He^{+} + He^{+}$ were measured. In this experiment it was found that an interference structure could be seen up to an impact energy of 1 MeV, which does not correspond to the adiabatic region anymore. To observe a significant structure in the MO spectra one should probably not use a collision energy higher than 50 MeV/u for the U^{92+}-Pb system, which is far below the relativistic matching energy of uranium of 240 MeV/u [12]. The reason that here the interference structure disappears much earlier is the additional time dependence in the occupation of the molecular orbitals. Calculations in which we included only the $1s\sigma$ and $2p\sigma$ orbitals and where we kept the occupation of these constant lead to an interference structure even for the highest taken impact energy of 100 MeV/u [13].

In Fig. 4 the emission spectra for the impact parameters of 20 fm and 50 fm are compared for three different collision energies. One can see a very strong dependence on the impact parameter for low impact energies, but almost no dependence for the impact energy of 100 MeV/u. This has to be explained in the same context as the impact energy dependence. The interference structure appears only for slow collisions and is in a first approximation determined by the phase factors entering the transition probability [4]. However, these depend on the impact parameter, which explains the shift in the structure for changing impact parameter.

In systematic investigations of the effect of rotational and radial coupling on the spectra we have found that both have to be considered. The MO x rays act as a sensitive probe on the occupation (especially at avoided crossings) of the molecular orbitals and neglect of either one of the couplings is seen in the spectra immediately [13].

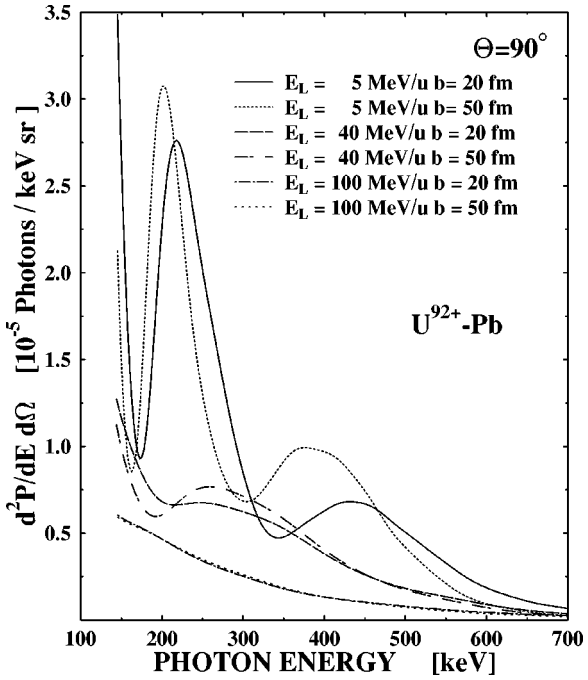

FIG. 4. Double differential emission spectra from a U^{92+}-Pb collision for three different impact energies and the impact parameter $b=20$ and 50 fm. The photon is emitted at an angle of $\Theta=90°$ with respect to the beam axis in the laboratory system.

With these results we are able to show that the investigation of quasimolecular MO x rays in collisions with H-like or fully stripped projectiles is a very powerful tool to investigate the transiently formed molecular orbitals. Measuring these spectra would allow a further approach to get information of the $1s\sigma$ orbital of superheavy quasimolecules. We have calculated spectra for the U^{92+}-Pb collision system and analyzed their structure for varying impact energies. Our results should be taken into account for an experimental realization since we found that the striking features in the spectra are only seen in a small range of impact energy.

K.S. and J.A. gratefully acknowledge the financial support from the Gesellschaft für Schwerionenforschung.

[1] I. Tserruya, R. Schuch, H. Schmidt-Böcking, J. Barrette, Wang Da-Hai, B. M. Johnson, M. Meron, and K. W. Jones, Phys. Rev. Lett. **50**, 30 (1983).

[2] R. Schuch, H. Schmidt-Böcking, I. Tserruya, B. M. Johnson, K. W. Jones, and M. Meron, Z. Phys. A **320**, 185 (1985).

[3] R. Schuch, M. Meron, B. M. Johnson, K. W. Jones, R. Hoffmann, H. Schmidt-Böcking, and I. Tserruya, Phys. Rev. A **37**, 3313 (1988).

[4] K. Schulze, J. Anton, W.-D. Sepp, P. Kürpick, and B. Fricke, J. Phys. B **30**, L67 (1997).

[5] U. Müller-Nehler and G. Soff, Phys. Rep. **246**, 101 (1994).

[6] J. Kirsch, B. Müller, and W. Greiner, Z. Phys. D **1**, 47 (1986).

[7] G. Wintermeyer, V. Dangendorf, H. Schmidt-Böcking, T. Kambara, P. H. Mokler, R. Schuch, and I. Tserruya, Z. Phys. D **17**, 145 (1990).

[8] J. Kirsch, W. Betz, J. Reinhardt, B. Müller, W. Greiner, and G. Soff, Z. Phys. A **292**, 227 (1979).

[9] J. Kirsch, B. Müller, and W. Greiner, Z. Naturforsch. A **35A**, 579 (1980).

[10] J. Anton, K. Schulze, P. Kürpick, W.-D. Sepp, and B. Fricke, Hyperfine Interact. **108**, 89 (1997).

[11] V. Mergel, R. Dörner, J. Ullrich, O. Jagutzki, S. Lencinas, S. Nüttgens, L. Spielberger, M. Unverzagt, C. L. Cocke, R. E. Olson, M. Schulz, U. Buck, and H. Schmidt-Böcking, Nucl. Instrum. Methods Phys. Res. B **98**, 593 (1995).

[12] J. Eichler and W. E. Meyerhof, *Relativistic Atomic Collisions* (Academic, San Diego, 1995).

[13] K. Schulze *et al.* (unpublished).