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# Study of $\delta$ -electron spectra produced during the collisions of Br on Pb, Br on U, and I on Pb

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Differential cross sections  $d^2\sigma/dE_e d\Omega_e$  of the  $\delta$ -electron emission from superheavy quasiatoms formed during heavy-ion collision of the systems Br on Pb and Br on U were measured for beam energies of 3.88 and 4.66 MeV/u. These cross sections were compared with the available theoretical results. Binding-energy differences of K- and L-shell electrons of the quasiatoms are extracted from the differential cross sections and compared with theoretical values. The measured angular distribution of  $\delta$  electrons emitted from the collision system I on Pb at a beam energy of 4.0 MeV/u shows a clear isotropic behavior.

## I. INTRODUCTION

The  $\delta$ -electron spectroscopy has been successfully used<sup>1-3</sup> for the study of the inner-shell ionization of super-heavy quasiatoms formed during a heavy-ion collision. It has also been shown<sup>3</sup> that it is possible to evaluate the inner-shell electron binding energies of the quasiatoms from the measured electron-emission cross sections. The theoretical values are in good agreement with these experimental results.

In this paper we present the experimental doubledifferential cross sections  $d^2\sigma/dE_e d\Omega_e$  of  $\delta$ -electron emission of the systems Br on Pb and Br on U. The corresponding united atomic charge  $Z_u$  of these systems are 117 and 127, respectively. The basic measurements were electron singles spectra and the spectrum of the electrons in coincidence with the characteristic K x rays of the heavier collision partner. Since the decay time of the K vacancy ( $\sim 10^{-17}$  sec) is much greater than the collision time (~ $10^{-20}$  sec) the decay of the K vacancy takes place long after the collision. Therefore the coincidences between electrons and K x rays of the heavier partner give the electrons emitted from the K shell of the quasiatom. The experimental setup and the techniques used are briefly described in Sec. II. We have been able to measure the electron energies up to 750 keV. The electron spectra show that high momenta are transferred only to the strongly bound inner-shell electrons. The results and the extracted binding-energy differences of K- and L-shell electrons are shown in Sec. III. Finally the results of angular-distribution measurements of  $\delta$  electrons are presented. Such results support the validity of the widely used monopole approximation<sup>4</sup> for heavy systems.

## **II. EXPERIMENTAL SETUP AND TECHNIQUES**

The experiments were performed at the Heidelberg MP Tandem Van de Graaff accelerater in combination with the post accelerator using beams of Br and I. The projectile energies were chosen to be below the Coulomb barrier to avoid background due to nuclear reactions. The targets were self-supporting Pb and U foils of thicknesses 0.5 to  $1 \text{ mg/cm}^2$ .

The experimental setup consisted of two electron detectors, two NaI(Tl) detectors to detect x rays, and a surface barrier Si(Li) detector for monitoring the beam. To measure the electrons away from the target in a backgroundsuppressed area, a magnetic transport system<sup>5</sup> having two identical achromatic electron channels is used. Each of the electron channels has four vertical dipole magnets made from soft iron wound with copper wires. After passing through these achromatic channels, electrons are detected by two Si(Li) detectors. The solid angle of each channel was measured to be 21.2 msr. The electrons are selected according to their momenta by a slit system. The momentum acceptance (byte) with this slit system was selected to be  $\Delta p/p = 32\%$ . The efficiency within the momentum byte is a constant. The energy resolution of the Si(Li) detectors is 2.5 keV at an energy of 1 MeV. One of the two 2 in.  $\times \frac{1}{4}$  in. x-ray detectors was mounted above the target and the other below the target such that they were in the same vertical plane to the beam direction. The total solid angle covered for x rays was 30% of  $4\pi$ .

# III. RESULTS

The Br on Pb system was studied with two different beam energies. The projectile energies were 3.88 and 4.66 MeV/u, respectively. These energies have been corrected for the energy loss in the target. The targets were selfsupporting Pb foils with thickness of 800  $\mu$ g/cm<sup>2</sup>. The singles and coincidence double-differential cross sections for this system with beam energy 3.88 MeV/u are shown in Fig. 1. An interesting feature of these spectra is the fast falloff of the cross sections with electron energy. The falloff of the total electron spectrum is more pronounced than the one for K-shell electron spectrum. For example, the ratio between these two cross sections at energies 80



FIG. 1. Double-differential cross sections  $d^2\sigma/dE_e d\Omega_e$  of total electrons and K-shell electrons of the system Br on Pb measured at beam energy of 3.88 MeV/u compared with theoretical calculations (Ref. 6).

and 500 keV are 117 and 2, respectively. The main contribution to the total electron spectrum stems from the Kand L shells. At low energies the total electron cross section is more than 2 orders-of-magnitude higher than the K-shell cross section. This means at low electron final energies the main contribution to the total electron spectrum comes from the L shell of the quasiatom. Therefore, the ratio at 80 keV is nearly the ratio between L- and Kelectron cross sections. At higher energies if the Kelectron contribution is subtracted from the total spectrum, then the ratio becomes even smaller than 2. This situation may be explained by considering the minimum momentum transfer to the electrons, which is given by  $q_0 = (E_B + E_f)/v_p^{cm}$ , where  $E_B$ ,  $E_f$ , and  $v_p^{cm}$  are the binding energy, the electron final energy, and projectile velocity, respectively. At  $E_f = 80$  keV the momentum transfer to the L electrons with binding energy 40 keV is equal to 1.8 MeV/c and that to K electrons with binding energy 200 keV is equal to 4.2 MeV/c. At  $E_f = 500$  keV the momentum transfers are 8.2 and 10.6 MeV/c, respectively. This means with a momentum transfer of 1.8 MeV/c, which is relatively low compared to 4.2 MeV/c, more electrons from the L shell can be removed than from the K shell because of their low L-shell binding energy. In this competition at low energies more electrons from L shell are emitted than from K shell. However, if one considers the high-energy electrons, the momentum transfer  $q \le q_0 = \Delta E / v_p^{cm}$  is large and is less affected by the difference in the binding energies of the L and K shell. Furthermore, since a high momentum can be effectively transferred only to the strongly bound electrons, more electrons are emitted from the K shell than from the Lshell if a fixed high-momentum transfer is considered.

The  $\delta$ -electron emission cross sections for a beam energy of 4.66 MeV/u are shown in Figs. 2 and 3. The electrons detected at 75° with respect to the beam axis are shown in Fig. 2. Figure 3 shows the cross sections of the electrons detected at 105°. The properties of these spectra are similar to that with a beam energy of 3.88 MeV/u. The cross sections obtained with higher beam energy are larger than with lower beam energy. For the beam energy of 4.66 MeV/u the cross section at  $E_f = 300$  keV is a factor of 1.8 larger than that of 3.88 MeV/u. Since Pb is a spherical nucleus, within the energy region of interest there is no contribution from conversion electron lines originating from Pb excitations. However, at ~520 keV there is a broad bump (see Fig. 1) which can be attributed to the internal conversion of the transition from  $\frac{5}{2}^{-1}$  state to  $\frac{3}{2}^{-1}$  in Br.

The solid lines in Fig. 1 show the results of coupledchannel calculations of de Reus et al.<sup>6,7</sup> They have obtained these results solving the stationary two-center Dirac equation in the monopole approximation, including corrections for electron-electron interaction in the Hartree-Fock-Slater approximation, and performing a coupled-channel calculation for the electron excitation amplitudes. As can be seen, the theoretical calculation of both the K shell and total electron-emission cross sections agree rather well with our experimental results. Since the monopole approximation is used in these calculations the theoretical results are independent of the electron angle. The corresponding theoretical results for the higher beam energy are shown in Fig. 3 as the solid curves are the results of coupled-channel calculations. The dashed lines in Fig. 2 show the calculation of Jakubasa. In contrast to the coupled-channel calculations of Refs. 6 and 7, this calculation was performed using perturbation theory,<sup>8</sup> with an effective united atomic charge which is adjusted to the total ionization cross section. As can be seen, the results of Jakubasa for total electron emission agree well with our experimental results. In these calculations only the contribution from L and K shells have been taken into account. This also suggests that the contribution to the total electron spectrum is predominantly from the K and L shells<sup>3</sup> of the quasiatoms. However, Jakubasa's results for Kshell electrons deviate from the experimental results. At very low electron energy the results agree fairly well. But at higher energies theoretical results overestimate the cross section by a factor of 2 at  $E_f = 500$  keV. Surprisingly, for higher beam energy (4.66 MeV/u) both theoretical results show a similar deviation from experiment at higher electron energies. A remarkable difference is that at low projectile energy (3.88 MeV/u) coupled-channel



FIG. 2. Double-differential cross sections  $d^2\sigma/dE_e d\Omega_e$  of total electrons and K-shell electrons of the system Br on Pb measured at beam energy of 4.66 MeV/u and electron angle 75° compared with theoretical calculations (Ref. 8).



FIG. 3. Double-differential cross sections  $d^2\sigma/dE_e d\Omega_e$  of total electrons and K-shell electrons of the system Br on Pb measured at beam energy of 4.66 MeV/u and electron angle of 105° compared with theoretical calculations (Ref. 6).

calculations agree with the experimental results, but at a higher projectile energy of 4.66 MeV/u, theory slightly overestimates the results.

A second system, Br on U, was also studied for two different beam energies 3.88 and 4.66 MeV/u, respectively. The targets were self-supporting U foils of thickness 1 mg/cm<sup>2</sup>. For this system, with lower beam energy, electrons were measured up to  $\sim 620$  keV. The cross sections with lower beam energy are shown in Fig. 4 for an emission angle of 105°. The results with higher beam energy are shown in Fig. 5. In this case electron energies were measured up to  $\sim$ 750 keV. The characteristic features of these cross sections are similar to those of Br on Pb. The only remarkable difference is the number of conversion electron lines in the spectra. These lines have been identified as due to Coulomb excitation of the ground-state rotational band in <sup>238</sup>U. This is one of the problems encountered when systems involving deformed nuclei are studied. The solid lines in Figs. 4 and 5 are the recent results of coupled-channel calculations of Mehler.<sup>7</sup> At low beam



FIG. 4. Double-differential cross sections  $d^2\sigma/dE_e d\Omega_e$  of total electrons and K-shell electrons of the system Br on U measured at beam energy of 3.88 MeV/u and electron angle of 105°. The solid lines are the theoretical calculations of Mehler *et al.* (Ref. 7). The arrows indicate the conversion electron lines due to Coulomb excitation of the ground-state rotational band of <sup>238</sup>U.



FIG. 5. Double-differential cross sections  $d^2\sigma/dE_e d\Omega_e$  of total electrons and K-shell electrons of the system Br on U measured at beam energy of 4.66 MeV/u and electron angle of 105°. The solid lines are the theoretical calculations of Mehler *et al.* (Ref. 7). The arrows indicate the conversion electron lines due to Coulomb excitation of the ground-state rotational band of <sup>238</sup>U.

energy (3.88 MeV/u) again the theory agrees fairly well with our experimental results. However, in the case of 4.66 MeV/u, theoretical results slightly overestimate the cross sections. For instance, in the case of total electrons these calculations overestimate the cross section by a factor of 1.4 at  $E_f$ =400 keV.

The double-differental cross sections of electrons (for a given electron energy) of the system Br on U is distinctly higher compared with the system Br on Pb. For example, for the beam energy of 3.88 MeV/u, the ratio of K-shell electron cross section of Br on U to Br on Pb at  $E_f = 300$  keV is 1.8. This and other results<sup>5</sup> show clearly an increase in cross section with an increase of  $Z_u$ , which is due to the shrinking<sup>9,10</sup> of the electron wave function (increase of electron density) with increasing  $Z_u$ .

#### **IV. BINDING ENERGIES**

From the first-order time-dependent perturbation theory the emission amplitude for electrons from an initial state i to a final state f is given by<sup>11</sup>

$$a(t) = -\int_{-\infty}^{+\infty} dt \, \dot{R}(t) \left\langle \phi_f \left| \frac{\partial}{\partial R} \right| \phi_i \right\rangle \\ \times \exp\left[ i \int_{-\infty}^{t} (E_f - E_i) dt' \right]$$

using the identity<sup>12</sup>

$$(E_f - E_i) \left\langle \phi_f \left| \frac{\partial}{\partial R} \right| \phi_i \right\rangle = \left\langle \phi_f \left| \frac{\partial V}{\partial R} \right| \phi_i \right\rangle$$

and for the symmetric Rutherford trajectory this reduces to

$$a(t) = 2i \int_{-\infty}^{+\infty} \frac{dR}{\Delta E} \left\langle \phi_f \left| \frac{\partial V}{\partial R} \right| \phi_i \right\rangle \sin(\Delta E t) ,$$

where V and R are the two-center potential and distance of the target and projectile system, respectively, and

$$\Delta E = |E_B| + E_f$$

The probability of electron emission from an initial state i is then

$$P_i(R,\Delta E) \sim \left| \int_{R_0}^{\infty} \frac{dR}{\Delta E} M_i(R) \sin(\Delta E t) \right|^2$$

where the matrix element  $M_i = \langle \phi_f | \partial V / \partial R | \phi_i \rangle$  depends only on R and the electron final energy  $E_f$ . If the monopole approximation<sup>4</sup> is assumed then for  $r_e > R/2$ ,  $\partial V / \partial R = 0$ . This means the major contribution to the emission amplitude comes when the nuclei are at closest distance, as it is necessary only to consider the small R region. The wave functions of K and L shell have nearly the same asymptotic behavior<sup>3</sup> for  $r_e \rightarrow 0$ .

This gives

$$\frac{|\phi_{1s_{1/2}}(r_e \to 0)|^2}{|\phi_{2s_{1/2}}(r_e \to 0)|^2 + |\phi_{2p_{1/2}}(r_e \to 0)|^2} = \text{const}$$

For a fixed  $\Delta E$ , i.e.,  $\Delta E = \Delta E_K = \Delta E_L$ ,

$$P_K(R,\Delta E) \sim \left| \int_{R_0}^{\infty} \frac{dR}{\Delta E} M_{1s_{1/2}}(R) \sin(\Delta E t) \right|^2$$

and

$$P_L(R,\Delta E) \sim \left| \int_{R_0}^{\infty} \frac{dR}{\Delta E} M_{2s_{1/2}}(R) \sin(\Delta E t) \right|^2 + \left| \int_{R_0}^{\infty} \frac{dR}{\Delta E} M_{2p_{1/2}}(R) \sin(\Delta E t) \right|^2$$

Since the R dependence of the K- and L-shell electronic wave functions are the same, the R dependence of the matrix elements are also the same. Therefore, if the matrix elements are corrected for the final electron energy dependence, the following result is readily obtained:

$$\frac{\sigma_K}{\sigma_L} = \frac{|M_{1s_{1/2}}|^2}{|M_{2s_{1/2}}|^2 + |M_{2p_{1/2}}|^2} = C(\text{const}),$$

where  $\sigma_K$  and  $\sigma_L$  are the cross sections for the K and L shells, respectively.

Therefore for a given momentum transfer  $q_0$  (or a given  $E_f + E_B$ , the ratio between K- and L-shell electron cross sections is given by the ratio between the squares of the respective transition matrix elements. This ratio is a constant for a given projectile and target system and varies smoothly with the electron final energy. After they were corrected for the final energy dependence, the values obtained for this constant C for Br on Pb and Br on U systems are 3.2 and 2.7, respectively. This means that for given energy transfer the double-differential cross section of K-shell electrons must be larger than that of L-shell electrons and one has to shift the K-electron spectrum (Figs. 1-5) along the energy axis in order to compare the K- and L-shell electron cross sections at the same energy transfer. For the same energy transfer  $(\Delta E_L = \Delta E_K)$  to both K- and L-shell electrons one gets

$$E_B^K - E_B^L = E_f^L - E_f^K$$

Therefore at the same energy transfer to the electrons

TABLE I. Binding-energy difference between K- and L-shell electrons of quasiatoms with united atomic charges 117 and 127 compared with theoretical predictions.

$Z_u$	$\Delta E_B \text{ (theory)} \\ \text{(Ref. 13)}$	$\Delta E_B \text{ (theory)} \\ \text{(Ref. 14)}$	$\Delta E_B$ (experiment)
117	172	171	162±12
127	213	216	191±16

the binding-energy difference of K- and L-shell electrons is given by the final energy difference (shift) of the K- and L-shell electrons. By subtracting the K-shell spectrum from the total spectrum, the electron spectrum for the Lshell can be obtained. The comparison of electron spectra can be made in the following way. First a value for the electron final energy is chosen. The corresponding Kshell cross section is read in the K-shell spectrum. Then the K-shell spectrum is shifted along the energy axis until the cross-section ratio is equal to the value given by the matrix elements. The binding-energy difference obtained from this method for the above systems are tabulated in Table I. In the trajectory of the projectile and target nuclei, electrons are mainly emitted near the distance of closest approach. Therefore, what one measures experimentally are the electron spectra and thus the binding energies of the quasimolecule formed by the target and projectile nuclei near the distance of closest approach.

Theoretical binding-energy differences<sup>13,14</sup> for these systems are also tabulated in Table I. These theoretical results are in good agreement with our experimental results. However, for the higher united atomic charges the theoretical values are slightly higher than the experimental ones.



FIG. 6. Probability of electron emission plotted against distance of closest approach  $R_0$  for electron-emission angles 62°, 80°, 105°, and 122° and electron final energies of 132 and 234 keV.



FIG. 7. Probability of electron emission plotted against both electron-emission angle  $\theta_e$  and the relative angle  $\theta_{e,HI}$  between  $\theta_e$  and the heavy-ion scattering angle for electron energy of 132 keV.

## V. ANGULAR DISTRIBUTION OF δ ELECTRONS

For the measurements of angular distribution the I on Pb system, which has a united atomic charge of 135, was used. The target was a self-supporting Pb foil with a thickness of 1 mg/cm<sup>2</sup> and the beam energy was 4.0 MeV/u. The experimental setup used in this case is described in Ref. 3. Scattered heavy ions were measured in addition to the electrons and x rays using a position sensitive avalanche counter with eight concentric rings as anode and a polycarbonate foil covered on one side with an evaporated gold layer of 0.2  $mg/cm^2$  as cathode. The foil was subdivided into eight sectors. The counter in this way was position sensitive to nuclear scattering angles of 10° to 40° and to all azimuthal angles  $2\pi$ . In order to evaluate the emission probability as a function of the distance of closest approach  $R_0$ , coincidence between electrons and scattered heavy ions were measured. These measurements were taken for four different electron emission angles  $\theta_{e}$  and  $\theta_{e}$  was measured with respect to the beam direction. Coincidences between scattered projectiles and electrons were separated by settting the windows on energy loss spectra of the scattered heavy ions. In recent measurements projectiles and recoils were separated by measuring<sup>5</sup> the time of flight of projectiles and recoils.



FIG. 8. Probability of electron emission plotted against both electron-emission angle  $\theta_e$  and the relative angle  $\theta_{e,HI}$  between  $\theta_e$  and the heavy-ion scattering angle for electron energy of 234 keV.

The probability of production of delta electrons as a function of distance of closest approach  $R_0$  is shown in Fig. 6. The lower set of data points corresponds to an electron mean final energy of 132 keV and the upper set of data points corresponds to that of 234 keV in the c.m. system.  $P(R_0)$  normalized to its average over electronemission angles  $\theta_e$  is shown as a function of  $\theta_e$  in Figs. 7 and 8. The eight sets of points correspond to eight rings, which gives the scattering angles of heavy ions, and each set consists of four points corresponding to four angles of electron emission as shown in Figs. 7 and 8 with opened and closed circles. The other points are the emission probability obtained from the sector information as described in the following.

Because of the symmetry of sectors with respect to the electron-emission angle, the data from eight sectors were reduced to four sectors by adding the data from the opposite sectors. Each sector which defines a unique azimuthal angle  $\phi$  is further divided into eight segments with a concentric ring counter placed behind the foil. Each ring defines a unique heavy-ion scattering angle  $\theta$ . These give rise to 32 different directions  $(\theta, \phi)$  and these directions with respect to the electron-emission angle  $\theta_e$  is defined as  $\theta_{e,HI}$ . The electrons measured in coincidence with heavy ions scattered to these segments were normalized to corre-

sponding heavy-ion singles and finally to  $P(R_0)$ . The results are shown in Figs. 7 and 8.

According to the above results, Figs. 6–8, the angular distribution of the  $\delta$  electrons emitted from superheavy quasiatoms, show an isotropic behavior within the range of angles measured in this experiment. However, one can see from these results that one cannot rule out anisotropies of about 10% or less. This implies that the monopole approximation<sup>4</sup> is a good approximation to describe the  $\delta$ -electron emission process in superheavy quasiatoms. However, this can depend on the system considered, for example, the dipole contribution is zero in symmetric systems.<sup>15</sup> Certainly higher-order multipoles contribute in this process. However, the contribution is smaller for heavy systems. Also, measurements<sup>16</sup> with lighter systems do not show a similar isotropy.

# **VI. CONCLUSIONS**

From the characteristics of the cross sections we have seen that high momenta can be transferred only to strongly bound inner-shell electrons. The increase of electronemission cross section for a given minimum momentum transfer with the increase of united atomic charge indicate the increase of electron density during the collision. In the case of Br on Pb at lower beam energies, coupledchannel calculations as well as the perturbation theory, working with an effective united atomic charge, reproduces the cross sections. However, at higher beam energies and at high electron energies, theoretical results slightly overestimate the cross sections. The theoretical binding-energy difference of K- and L-shell electrons with lower atomic number agrees reasonably with our experimental results. Within the limits of statistical and systematical errors (~10%) the angular distribution of  $\delta$ electrons of heavy systems was found to be isotropic, supporting the validity of the monopole approximation.

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