

Evidence for an additional coupling of the innermost shells in very heavy quasi-molecular ion–atom collisions

M Mann[†], P H Mokler[‡], B Fricke[†], W-D Sepp[†], W A Schönfeldt[‡], and H Hartung[†]

[†] Fachbereich Physik, University of Kassel, West Germany

[‡] Gesellschaft für Schwerionenforschung, Darmstadt, West Germany

Received 25 June 1982

Abstract. Due to the tremendous spin-orbit splitting of quasi-molecular levels in super-heavy collision systems ($Z = Z_1 + Z_2 \geq 137$) bombarding energy $0.5\text{--}6 \text{ MeV } N^{-1}$, unusual couplings may occur around $Z = 165$. Experimental evidence for such a theoretically predicted coupling is discussed.

For very heavy collision systems with $Z = Z_1 + Z_2 \geq 100$, systematic total and differential cross section measurements for inner-shell excitation have been reported for quasi-adiabatic collision velocities (Behncke *et al* 1980, Anholt *et al* 1979, Meyerhof *et al* 1976, 1977, Liesen *et al* 1980b). For a summary of the experimental data see, e.g. Liesen *et al* (1980a), Mokler *et al* (1981), Mokler and Liesen (1982). The inner-shell excitation in this domain can be described reasonably well by theories where the monopole approximation is used for the quasi-molecular levels as well as the interaction (Müller and Greiner 1976, Betz *et al* 1976, Soff *et al* 1977, 1980, Reinhardt *et al* 1979, Bosch *et al* 1980). Nevertheless, there are points, distinctly open, which need clarification. The aim of this article is to discuss one of these points in the framework of new multielectron correlation diagrams and coupled channel calculations for the heaviest collision systems. We will concentrate on the $2p_{1/2,1/2}$ excitation, where, as a function of $Z_1 + Z_2$ around 164, an enhancement in the total cross section is visible (Behncke *et al* 1980, Anholt *et al* 1979, Mokler *et al* 1981, Mokler and Liesen 1982). (In non-relativistic cases the $2p_{1/2,1/2}$ level is called the $2p\sigma$ level.)

$2p_{1/2,1/2}$ excitation cross sections (Behncke *et al* 1980, Anholt *et al* 1979, Meyerhof *et al* 1976, 1977) are summarised for I, Xe, Pb, and U projectiles on various elements Z_2 at different bombarding energies from 0.5 to $6 \text{ MeV } N^{-1}$ in figure 1. The cross sections are plotted as a function of the atomic number of the united atom (UA) for $Z \geq 100$. Neglecting normalisation problems for the data sets of the various projectiles we find: for $Z \leq 145$ the $2p_{1/2,1/2}$ cross sections decrease roughly exponentially with Z ; whereas around $Z \approx 165$ a broad, peak-like cross section enhancement is observed which is most pronounced at low impact energies (1.4 to $3.6 \text{ MeV } N^{-1}$). At higher impact energies it gets broader and tends to vanish. Despite the experimental uncertainties in the data, one can conclude that the position of the hump structure depends on Z only and not on the asymmetry of the collision system (e.g., $|Z_1 - Z_2|/Z$). Hence, we may call this cross section enhancement a Z resonance.

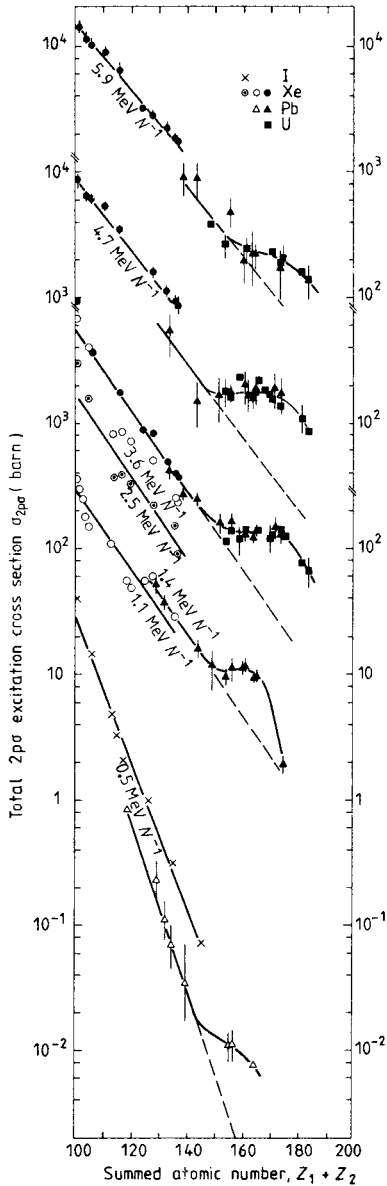


Figure 1. Experimental $2p_{1/2,1/2}$ excitation cross section for superheavy collision systems, $(Z_1 + Z_2) \geq 100$ for 0.5 to 6 MeV N^{-1} . (Behncke *et al* 1980, Anholt *et al* 1979, Meyerhof *et al* 1976, 1977).

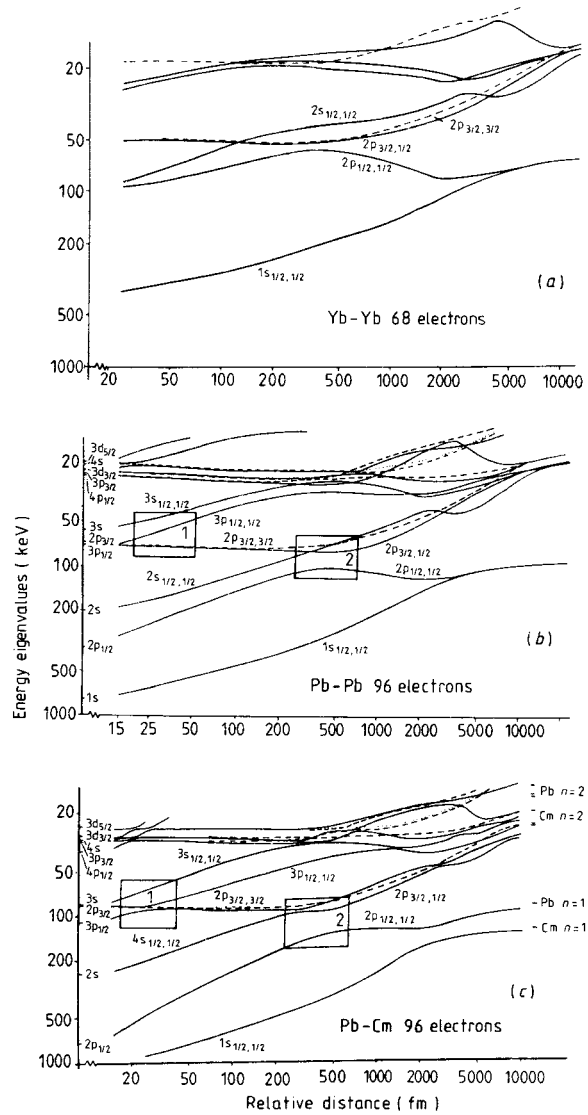


Figure 2. Calculated SCF-DFS multielectron correlation diagrams for: (a) Yb-Yb ($Z = 140$); (b) Pb-Pb ($Z = 164$); and (c) Pb-Cm ($Z = 178$).

It has been stressed already (Mokler *et al* 1981) that the almost exponential decrease in the $2p_{1/2,1/2}$ cross section with Z (for $Z \leq 145$) may indicate the dominance of the direct excitation of the $2p$ electrons in the quasi-molecule out of the demoted level in a single collision process. Due to the strong Z dependence of this process small multicollision contributions from projectile vacancies preformed in a solid target

which may be demoted from high-lying levels to the $2p_{1/2,1/2}$ level may finally contribute significantly to the cross section for higher Z .

The $2p_{1/2,1/2}$ cross section enhancement beyond $Z > 145$ was originally explained by relativistic effects: first-order perturbation theory gives the appropriate Z dependence in monopole approximation, but underestimates the experiments by a factor of four to five (Soff *et al* 1977, Anholt *et al* 1979). An inclusion in the calculations of so-called 'multistep processes' acting in one collision may increase the cross sections (Reinhardt *et al* 1979, Soff *et al* 1980). Cross section calculations performed so far are pure monopole approximations which include σ states and radial coupling, only. As fully dynamic calculations for the $2p_{1/2,1/2}$ excitation are not yet available, we will now try to interpret the observed Z resonance of the $2p_{1/2,1/2}$ cross section within a simple picture, where we correlate the cross section enhancement with isolated features in the corresponding correlation diagrams.

In figure 2 we have plotted correlation diagrams for $Z = 140, 164$, and 178 . The method of computation of these multielectron level diagrams has been reported elsewhere (Fricke *et al* 1976, Rosén and Ellis 1976). The diagram for the Pb-Pb system has been published already (Sepp *et al* 1981). Such multielectron calculations are needed in order to yield realistic level dependences with a correct screening. Compared with the one-electron calculations (Müller and Greiner 1976, Soff 1976) we find considerable deviations, in particular at larger internuclear distances.

Considering the correlation diagrams (figures 2(b) and 2(c)) we find that, due to the large spin-orbit splitting around $Z \geq 164$, the $3s_{1/2}$ and $3p_{1/2}$ levels from the UA M-shell approach the $2p_{3/2}$ level of the UA L shell. This is in contrast to lighter collision systems, see e.g. the correlation diagram for $Z = 140$ given in figure 2(a). Hence, for such heavy systems—like Pb-Pb ($Z = 164$)—performed, steady-state vacancies from the projectile M shell may have a chance to be transferred in the collision molecule at small internuclear distances into the $2p_{3/2}$ MO levels (i.e., $2p_{3/2,3/2}$, and $2p_{3/2,1/2}$). For the corresponding region see box 1 in figure 2. During the outgoing part of the collision these vacancies may cross at intermediate internuclear distances to the $2p_{1/2,1/2}$ level (see box 2 in figure 2). Summarising, an incoming projectile M-shell vacancy may end up finally as a $2p_{1/2,1/2}$ vacancy for the very heavy collision systems $Z \geq 164$.

For lighter collision systems, $Z \approx 140$, the reduction in spin-orbit splitting will decrease the coupling between the UA M and L shells (see box 1, figure 2). For heavier collision systems, $Z \approx 180$ the coupling strength between the relevant levels in box 2 may be reduced, due to the large spin-orbit splitting there. In addition for heavier collision systems the effective incoming M-shell ionisation will be reduced. The reasons are the following: for symmetric systems the M binding energy is increased; for asymmetric systems—if the projectile is the lighter particle—the M ionisation will be decoupled from the $3s_{1/2}$ and $3p_{1/2,1/2}$ levels; if the projectile is the heavier one the M ionisation is reduced due to the increased binding. Hence, around $Z \approx 164$ we may have a Z resonance-like enhancement for transferring preformed projectile M vacancies into the K shell of the lighter collision partner.

Assuming that around $Z \approx 160$ the cross section enhancement is caused primarily by a multicollision process (Liesen *et al* 1980b) which is determined by the projectile M pre-ionisation σ_{MC}^M , parameters for the relevant crossings can be extracted from existing data (Behncke *et al* 1980, Anholt *et al* 1979, Meyerhof *et al* 1976). For Pb-Pb collisions we find, for example, at $1.4 \text{ MeV } N^{-1}$, a cross section enhancement σ_{MC}^M of roughly 7 b which increases to about 100 b for the higher impact energies

around $4.7 \text{ MeV } N^{-1}$. These values can be taken as correct only within a factor of two, as they depend not only on the experimental uncertainties but additionally on the subtraction of the 'unknown' normal, i.e. single collision, cross section which was roughly extrapolated exponentially from lower Z values. (See broken curves in figure 1.)

In analogy to Meyerhof's description of the multicollision process in lighter collision systems (Liesen *et al* 1980b) the multicollision contribution to the inner-shell excitation cross section, caused by the projectile M-shell excitation, can be estimated by

$$\sigma_{MC}^M = g_M f_M \sigma_{\text{trans}}^M \bar{P}_L \quad (1)$$

where f_M is the average fraction of projectiles having a M vacancy, g_M is a factor giving the probability for the incoming vacancy to end up in the $3p_{1/2,1/2}$ (or $3s_{1/2,1/2}$) level, σ_{trans}^M is the transfer cross section for transferring this vacancy at small internuclear distances to the $2p_{3/2}$ MO levels (see box 1, figure 2) and finally, \bar{P}_L is the probability that this vacancy is additionally transferred at intermediate internuclear distances down to the $2p_{1/2,1/2}$ level on the outgoing part of the collision (see box 2 in figure 2).

The fraction f_M is given by

$$f_M = \sigma_M \rho L A^{-1} v \tau_M \quad (2)$$

where σ_M is the M ionisation cross section, $\rho L A^{-2}$ the number of target atoms per cm^3 , v the projectile velocity, and τ_M the lifetime of the projectile M vacancies. Using an excitation cross section σ_M of roughly 10^7 b for Pb–Pb collisions (Schönfeldt 1981) at $1.4 \text{ MeV } N^{-1}$ and using atomic values for τ_M we find fractions of about 30% for projectile M vacancies. Due to a probable, multiple M excitation (Schönfeldt 1981) this value has to be enlarged in reality. For g_M we will use only the normal, statistical factor 0.05 (or 0.11 including the $3s_{1/2,1/2}$ level), and neglect possible vacancy transfer to and from other levels due to couplings. Altogether, we get a $f_M g_M$ value for $1.4 \text{ MeV } N^{-1}$ of 0.01 to 0.03 as a minimum. Experimental cross sections σ_M for higher energies are not yet available, but it is known that the average charge state of a Pb projectile after traversing a solid target is 39 for $1.4 \text{ MeV } N^{-1}$ and 23, 52, and 58 for 0.5, 3.6, and $5.9 \text{ MeV } N^{-1}$, respectively (Franzke 1982). Similar values are expected within the solid target (Schönfeldt 1981). Since 55 is the charge state where, in an electronic ground state, the ionisation of the M shell starts, it is very probable that for the higher impact energies several M holes are present in every collision. Thus an estimate of the $f_M g_M$ value is somewhere about 0.5 for impact energies between 3.6 and $5.9 \text{ MeV } N^{-1}$. Further experiments have to yield better estimates for the $f_M g_M$ values.

In order to get quantitative numbers we have to estimate the transfer cross section $\sigma_{\text{trans}}^M P_L$ for the transfer of a $3p_{1/2,1/2}$ (and $3s_{1/2,1/2}$) hole into the $2p_{1/2,1/2}$ level. All our estimations are based on multichannel calculations using good correlation diagrams but model matrix elements, which are scaled from small Z crossings where the matrix elements are known. The results of the calculations are as follows. Strong rotational coupling, very similar to the normal $2p_\sigma$ – $2p_\pi$ coupling, transfers a large amount of the incoming holes to the $2p_{3/2,3/2}$ level. This level couples rotationally to the $2p_{3/2,1/2}$ and $2p_{1/2,1/2}$ levels which in turn couple radially. Finally, for one incoming $3p_{1/2,1/2}$ (or $3s_{1/2,1/2}$) vacancy, up to 0.5 holes may end up in the level of interest $2p_{1/2,1/2}$ for

impact parameters up to 150 fm. The total cross section is nearly constant at about 140 b from 1 to 6 MeV N^{-1} . If we multiply this with the above f_{MGM} value we get 3 to 6 b as the multicollision contribution to the cross section for 1.4 MeV and about 70 b for the higher energies. This is the correct order of magnitude.

The strong decrease of the multicollision contribution for higher Z systems, like Pb on U, is caused by decoupling of this channel from the incoming M shell. The coupling of the 3s and $3p_{1/2}$ united levels with the $2p_{3/2}$ level is in the same order of magnitude as in the Pb-Pb case (about 300 b for 4.7 MeV and 100 b for 1.4 MeV assuming one hole in the incoming $3p_{1/2}$ level). The total number of resulting $2p_{1/2,1/2}$ holes is decreased because here the 3s and $3p_{1/2}$ UA levels provide only a reduced fraction of vacancies compared with the lighter systems. For instance at 3.6 MeV N^{-1} the average charge state of U ions after penetrating a foil is 56 compared with 52 for Pb ions (Franzke 1982). However, we need charge states of 64 and 54 to open statically the U or Pb M shell, respectively. Hence for U-U collisions we expect a drastically reduced steady-state projectile M-shell excitation in the target compared with Pb-Pb collisions. As already mentioned at the beginning of this article corresponding arguments for an effective reduction in M vacancies transferred into the UA 3s and $3p_{1/2}$ levels at higher Z systems around 180 can be given as well. For instance for Pb on Cm we expect the 3s and $3p_{1/2}$ levels to be filled from the $n = 2$ levels of the higher Z partner which have less holes by two orders of magnitude. Thus the multicollision contribution cannot be much more than a few barns at higher energies also.

The same argument is true for the contribution of the incoming L shell in Pb-Pb via the direct rotational coupling from $2p_{3/2,3/2}$ to $2p_{1/2,1/2}$. The number of holes at the energies of interest will be at least two orders of magnitude smaller. As the cross section is expected to be somewhere around 500 b (for one hole in the $2p_{3/2,3/2}$ level) the contribution from this process will be only a few barns at the higher energies. A similar multistep rotational coupling mechanism was already postulated as the dominant $2p_{\sigma}$ excitation mechanism for U on Pb collisions and the general behaviour of the impact parameter dependence explained (Heiligenthal *et al* 1978). Nevertheless, the complicated behaviour of the impact parameter dependences of the $2p_{1/2,1/2}$ excitation found for different heavy systems (see Mokler *et al* 1981) is not understood at all. The multicollision contribution from the M shell to the inner-shell excitation should also manifest itself in the corresponding quasi-molecular radiation. The $2p$ MO x-ray yield should depend on the target thickness as the steady-state projectile M vacancies build themselves up in the first target layers. Looking at the large M-shell excitation cross sections we extract a mean free path for the projectile M-shell excitation in the region of $30 \mu\text{g cm}^{-2}$. Hence, we understand that beyond 1 mg cm^{-2} no target thickness dependence for the MO x-ray spectrum in Pb-Pb collisions was observed experimentally (Stoller *et al* 1980). Recent measurements with targets in the $100 \mu\text{g cm}^{-2}$ region show deviations from the early MO x-ray spectra (Stoller *et al* 1980) which can be understood in the picture of the discussed multicollision process (Stiebing 1982, Bethge *et al* 1981). Detailed investigations of the target thickness dependences for total and differential, non-characteristic and characteristic x-ray emission from inner shells can shed more light on the multicollision process discussed in this paper.

Summarising, we may say that the Z resonance which has been observed in heavy-ion collisions around $Z \approx 165$ can be explained by the unusual, additional coupling of the $3p_{1/2}$ and 3s UA levels which in this Z region are nearly degenerate

with the $2p_{3/2}$ UA level. Therefore, it is possible to transfer directly initial M holes around $Z \approx 165$ into the $1s$ level of the lighter atoms.

Moreover, we would like to mention that for the very heavy collision systems like Pb on Cm the spin-orbit splitting is so large that the $2p_{1/2,1/2}$ level tends to approach again the $1s_{1/2,1/2}$ level at very small internuclear distances, see the level diagram in figure 2(c). Due to the large spatial overlap of the corresponding wavefunctions a vacancy sharing between these levels may not only be active at large internuclear distances (Stoller *et al* 1980, Stiebing 1982, Bethge *et al* 1981) but additionally also at very small distances. Such a behaviour could possibly explain the increase in K-shell excitation probability of the heavier collision partner in the extremely heavy Pb on Cm collision system at small internuclear distances (see Liesen *et al* 1980a, b). Such a possibility has to be studied in more detail.

References

- Anholt R, Behncke H H, Hagmann S, Armbruster P, Folkman F. and Mokler P H 1979 *Z. Phys. A* **289** 349
- Behncke H H, Armbruster P, Folkman F, Hagman S, McDonald I R and Mokler P H 1979 *Z. Phys. A* **289** 333
- 1980 *Z. Phys. A* **294** 286
- Bethge K, Bosch F, Hagmann S, Liesen D, Maor D, Mokler P H, Schadt W, Schmidt-Böcking H, Schuch R, Stiebing K E and Vincent P 1981 *Report EAS-2* 68
- Betz W, Soff G, Müller B and Greiner W 1976 *Phys. Rev. Lett.* **37** 1046
- Bosch F, Liesen D, Armbruster P, Maor D, Mokler P H, Schmidt-Böcking H and Schuch R 1980 *Z. Phys. A* **296** 11
- Demkov Yu 1964 *Sov. Phys.-JETP* **18** 138
- Franzke B 1982 private communication
- Fricke B, Morović T, Sepp W D, Rosén A and Ellis D E 1976 *Phys. Lett.* **59** A 375
- Heiligenthal G, Betz W, Soff G, Müller B and Greiner W 1978 *Z. Phys. A* **285** 105
- Liesen D, Armbruster P, Behncke H H, Bosch F, Hagmann S, Mokler P H, Schmidt-Böcking H and Schuch R 1980a *Progress Report on XIth Conf. on Electronic and Atomic Collisions, Kyoto 1979* ed N Oda and K Takayunagi (Amsterdam: North-Holland) p 337
- Liesen D, Armbruster P, Bosch F, Hagmann S, Mokler P H, Schmidt-Böcking H, Schuch R, Wollersheim H J and Wilhelmy I B 1980b *Phys. Rev. Lett.* **44** 983
- Meyerhof W E 1973 *Phys. Rev. Lett.* **31** 1341
- Meyerhof W E, Anholt R and Saylor T K 1977 *Phys. Rev. A* **16** 169
- Meyerhof W E, Anholt R, Saylor T K, Lazarus S M, Little A and Chase L F 1976 *Phys. Rev. A* **14** 1653
- Mokler P H, Armbruster P, Bosch F, Liesen D, Maor D, Schönfeldt W A, Schmidt-Böcking H and Schuch R 1981 *Int. Conf. on X-ray Processes and Inner Shell Ionisation (X-80), Stirling 1980* (published in *X-ray Physics of Atoms and Solids* (New York: Plenum) p 49)
- Mokler P H and Liesen D 1982 *Progress in Atomic Spectroscopy Part C* ed H Kleinpoppen (New York: Plenum) to be published
- Müller B and Greiner W 1976 *Z. Naturf.* **31a** 1
- Reinhardt J, Müller B, Greiner W and Soff G 1979 *Phys. Rev. Lett.* **43** 1307
- Rosén A and Ellis D E 1976 *Phys. Lett.* **59A** 375
- Schönfeldt W A 1981 *Thesis* University of Köln (report GIS-81-7)
- Sepp W D, Fricke B and Morović T 1981 *Phys. Lett.* **81A** 258
- Soff G 1976 *Thesis* University of Frankfurt
- Soff G, Betz W, Heiligenthal G and Greiner W 1977 *Fizika* **9** Suppl. 4, 721
- Soff G, Reinhardt J, Müller B and Greiner W 1980 *Z. Phys. A* **294** 137
- Stiebing K E 1982 *Thesis* University of Frankfurt, to be published
- Stoller Ch, Morenzoni E, Wölfi W, Meyerhof W E, Folkman F, Vincent P, Mokler P H and Armbruster P 1980 *Z. Phys. A* **297** 93