

INTERPRETATION OF NONCHARACTERISTIC M X-RAYS IN HEAVY COLLIDING SYSTEMS BY SELFCONSISTENT RELATIVISTIC MOLECULAR CALCULATIONS*

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The result of the first calculation of a self-consistent relativistic many electron correlation diagram ever done (for the system Au-I) leads to a good agreement of the spectral shape and position of the observed noncharacteristic X-rays within the quasi adiabatic model.

The amount of experimental data of noncharacteristic molecular (MO) X-rays produced in heavy ion-atom collisions has very rapidly increased during the past few years [1]. The K MO X-rays appear as relatively structureless spectra exponentially decreasing with energy, which do not end at the united atom limit, but continue due to the dynamics of the collision process [2].

The M MO X-ray spectra observed so far in heavy ion-atom collisions like Au-I or U-I and partially the L MO X-rays like I-Ag instead show a significant structure [3]. The only hand-waving explanation up to now was that transitions into a level with a flat wide minimum in the unknown correlation diagram might generate the observed peak structure behaviour.

We were able to calculate the first relativistic many electron correlation diagram. Taking this result we show that a good interpretation of the observed structure is possible within the quasi-adiabatic model taking into account density of states arguments.

Recently an ab initio self-consistent relativistic Dirac-Slater molecular program for obtaining molecular energy levels and wavefunctions has been devel-

oped. In this program the molecular wavefunctions are expanded in linear combinations of basis functions which are chosen as numerical Dirac-Slater (DS) eigenfunctions of free atoms/ions located at given sites. DS calculations for systems [4] like XeF₂, InI, UO₂²⁺ as well as solids [5] showed the usefulness of this model. Details of the molecular computational method are given in ref. [6].

We have used this program to explore the collision process of the Au-I system which is the experimentally best known example for M MO X-rays. The resulting correlation diagram of this system taking 56 electrons into account is shown in fig. 1. As long as we do not have detailed experimental information on the actual degree of ionization and ionization distribution, the number of electrons chosen will be a relatively realistic assumption. We know from atomic calculations that the addition of further electrons has no significant effect on the transition energy for the M X-rays [7]. In molecular calculations one generally has to take into account relaxation effects and other binding energy corrections to the one-electron eigenvalues [8]. Here such effects are too small to be of relevance, compared to the uncertainties in the occupation numbers. It is clear that for sufficiently small internuclear distance R the elements of a two-center expansion basis

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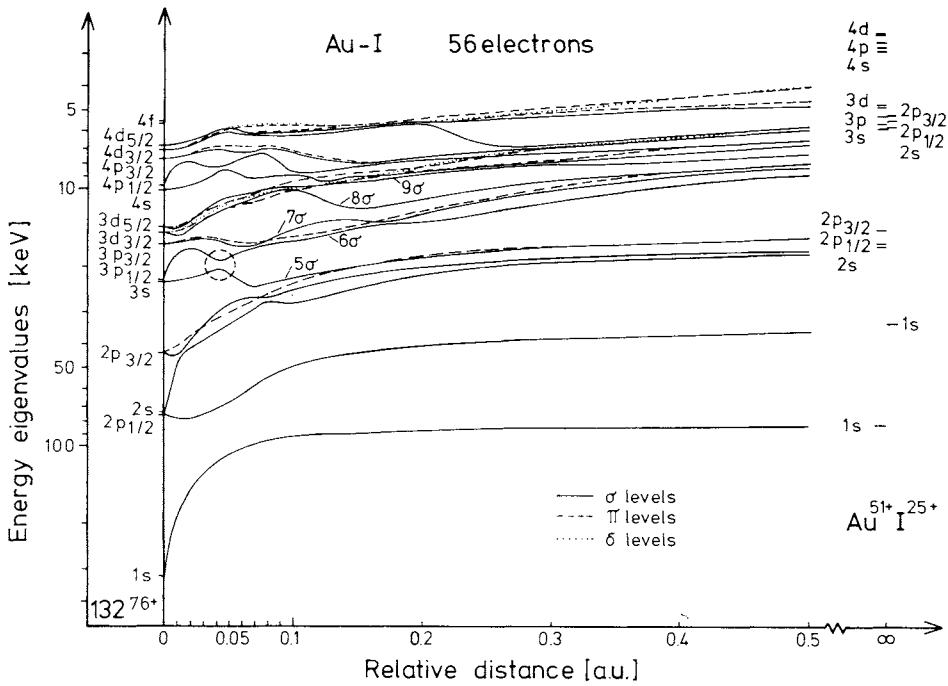


Fig. 1. Self-consistent relativistic correlation diagram of the system Au-I including 56 electrons. The relevant levels for the MO X-rays are the 6σ , 7σ , 8σ and 2π levels. Through the encircled crossing the characteristic Au L X-rays are feeded.

may become linearly dependent and precautions must be taken to avoid difficulties. In the present calculations this problem appeared for $R \lesssim 0.04$ au. Matrix reduction procedures based on Löwdin's canonical orthogonalisation procedure [9] or the use of a one center basis expansion would be equally effective solutions to this problem. In our case it was decided to connect the MO levels with united atom levels as a sufficiently accurate procedure.

From the MO level diagram shown in fig. 1 we have extracted all transition energies ΔE as function of R within the energetical range 5.5–11 keV where the MO peak appears as shown in fig. 2.

The most appropriate way to construct the predicted spectra at this stage is to introduce the transition density,

$$N(E, R) = \sum_{i < j} g_i g_j f_i (1 - f_j) P(\Delta E_{ij}) \frac{\sigma/\pi}{(E - \Delta E_{ij})^2 + \sigma^2} \quad (1)$$

Here $N(E, R)$ is a measure of the spontaneous emission at energy E for a given internuclear separation R . The factors g_i and f_i are orbital degeneracies and occupation probabilities respectively, σ represents the collision induced lifetime broadening of the $i \rightarrow j$ transition, and $P(\Delta E_{ij})$ describes the transition strength according to the multipole selection rules of the $i \rightarrow j$ transition. We can sum these transitions over all R

$$N(E) = \int_{R_{\min}}^{\infty} dR W(R) N(E, R) \Big/ \int_{R_{\min}}^{\infty} dR W(R) \quad (2)$$

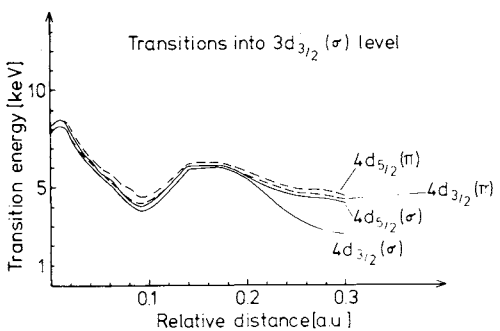


Fig. 2. Transition energies ΔE_{ij} versus internuclear distance R for the four levels relevant for the M MO X-rays.

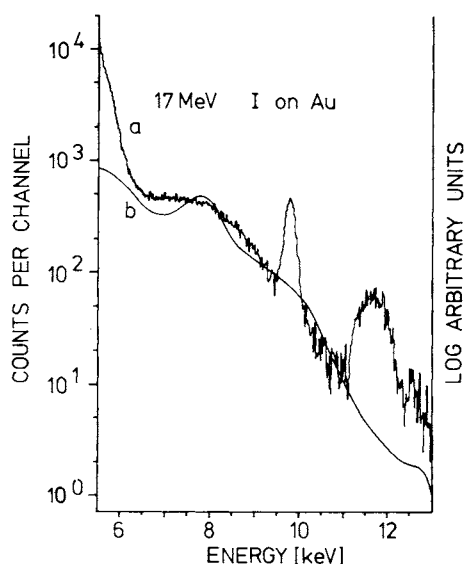


Fig. 3. X-ray spectrum for 17 MeV I on Au: (a) Measured spectrum corrected for absorber (refers to the left ordinate). (b) Calculated unnormalized MO-spectrum taking into account all transitions shown in fig. 2 with equal occupation probabilities (refers to the right ordinate).

with a weight factor $W(R)$ proportional to the fraction of time spent around distance R for every possible Coulomb trajectory at given impact energy E_{ion} . If we assume classical Rutherford trajectories, expression (2) can be exactly integrated over all impact parameters [10]. This leads to

$$N(E) = \frac{4\pi}{\sqrt{V(R_{\text{min}})}} \times \int_{R_{\text{min}}}^{\infty} dR R^2 \sqrt{1 - \frac{V(R)}{V(R_{\text{min}})}} N(E, R) \quad (3)$$

where R_{min} is the distance of closest approach for a head on collision for a given impact energy, $V(R)$ is the potential and μ reduced mass of the system. If we assume as a first approximation that all four relevant levels (see fig. 2) have the same number of holes, take $\sigma = 0.25$ keV, and assume that all transitions have dipole character which roughly leads to a $(\Delta E_{ij})^3$ behav-

our [11] for $P(\Delta E_{ij})$, we get the spectrum (b) shown in fig. 3 for 17 MeV projectile energy of I on Au. This peak agrees rather well with the experimental peak position. Calculations for different impact energies E_{ion} lead to spectra with the same center of gravity, becoming somewhat broader with increasing E_{ion} . This result shows that the peak structure behaviour is a direct result of the nearly constant behaviour of the transition energies of fig. 2 around 7.5 keV between $R \approx 0.11$ and 0.19 au.

Furthermore one can see that the character of the level crossings involving the 5σ to 9σ levels and thus their occupation numbers are most important. The intensity of the MO peak relative to the characteristic L X-rays will be strongly dependent on the character of the level crossing at 0.05 au (see circle in fig. 1) which seems to be relatively diabatic. Lutz et al. [12] have shown that with decreasing projectile energy the characteristic Au L X-ray line disappears at exactly the point where the energy becomes too small to reach this avoided crossing at 0.05 au at which the transfer of a hole into the L shell of the heavier atom is possible. But at this energy the MO X-ray peak can still be seen showing that the 6th–8th σ levels still carry a considerable number of holes. The MO peak disappears exactly at the point where the projectile energy becomes too small to reach the avoided crossing between the 8σ and 9σ level near 0.1 au so that no holes can be promoted down into the 8σ level anymore. These results strongly indicate that the calculated correlation diagram is very realistic allowing such a detailed interpretation even for these high levels.

The shape of the spectrum will of course also be dependent on the number of holes created in these levels during the collision and their dependence on the impact energies. A detailed discussion of these dependencies can only be given if we know the relevant ionization processes. Such data are beginning to be available from gas target experiments [13].

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