PHYSICAL REVIEW A VOLUME 13, NUMBER 2 FEBRUARY 1976

Impact-parameter dependence of molecular orbital x rays produced by collisions of I with Au

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(Received 4 September 1974)

The coincidence spectra of molecular orbital x rays produced in the scattering of a beam of 26.85-MeV I ions by Au at laboratory angles of 1.5, 3.5, and 10.5 deg have been measured. Analysis of the data gives the energy dependence and approximate cross section for the molecular-orbital x rays for the combined system with $Z = 132$ as a function of impact parameter. The results confirm and extend the conclusions drawn from previous measurements of the x-ray singles spectrum.

I. INTRODUCTION

The x rays produced in the collisions of iodine beams with heavy targets such as Au, Th, and U have been studied in some detail recently.[1-3] In addition to the expected L and M x rays from the beam and target a broad band of x rays with energies between the iodine L x ray at ~4 keV and the target L x ray at ~10 keV was observed. In both experiments it was concluded that this x-ray band could be appropriately described as molecular-orbital (MO) M x rays from the quasiatom with $Z = 132$, 143, or 145 for the above targets. The measurement of the impact parameter dependence of MO x-ray production is an important experimental refinement, since such measurements make possible an accurate determination of the MO M-x-ray energy at small internuclear distances, an estimate of the spacing of the molecular orbitals as a function of internuclear distance, and from the cross sections and spectral shape an estimate of the transition probability along a given trajectory. The purpose of this paper is to demonstrate the usefulness and feasibility of coincidence experiments in the study of MO x rays.

II. EXPERIMENTAL TECHNIQUE AND RESULTS

A beam of 26.8-MeV I ions from the Strasbourg MP tandem Van de Graaff was used to bombard a 200-μg/cm^2 Au foil oriented at 45° to the beam axis. The beam was collimated to about ±0.06° by two 0.5-mm collimators 50 cm apart. The x rays emitted at 90° were detected with a cooled Si(Li) x-ray detector with a depth of 5 mm and an area of 80 mm^2, which subtended a calculated solid angle of 0.035 sr at the target. A preamplifier with pulsed optical feedback and a pileup rejection network was used. The resolution of the detector obtained for the iodine L x rays during the experiment was about 230 eV. The only absorber between target and detector was the 25-μm Be detector entrance window, which did not appreciably distort the spectrum in the x-ray energy range above 4 keV. Scattered particles where detected with a 100-μm-thick silicon surface barrier detector. An annular collimator with an inner diameter of 16 mm and a width of 0.5 mm was used to define the scattering angle. Variation of the scattering angle was obtained by changing the distance between target and collimated counter. Coincidences between events in the x-ray and particle detectors were obtained using standard electronics. The time resolution was about 250 nsec. The accidental coincidence rates were about 20% of the total coincidence rates for scattered particle rates of 3000–5000 sec^{-1}. Under these conditions the counting rate in the x-ray detector was ~200 sec^{-1} or less, which ensured that there were negligible problems from pileup. Coincidence measurements were made at laboratory angles of 1.5, 3.5, and 10.5°, which correspond to impact parameters of about 8.6, 3.7, and 1.2×10^{-10} cm and distances of closest approach of 8.8, 3.8, and 1.4×10^{-10} cm, respectively. The x-ray singles spectrum and the three coincidence spectra are shown in Fig. 1. An expanded view of the portion of the spectra of interest for MO x rays is shown in Fig. 2.

In the interpretation of the data the well defined endpoints of the coincidence spectra shown in Fig. 2 are assumed to be correlated with the impact parameter or distance of closest approach calculated for the appropriate scattering angle. We have done a set of calculations to investigate the possible influence on this assumption of the effect of multiple scattering as the beam traverses the foil as well as the additional effect of a *possible* two-step reaction mechanism.[1-3] The *actual* reaction mechanism must be regarded at the present time as not demonstrated experimentally in a conclusive or convincing manner.

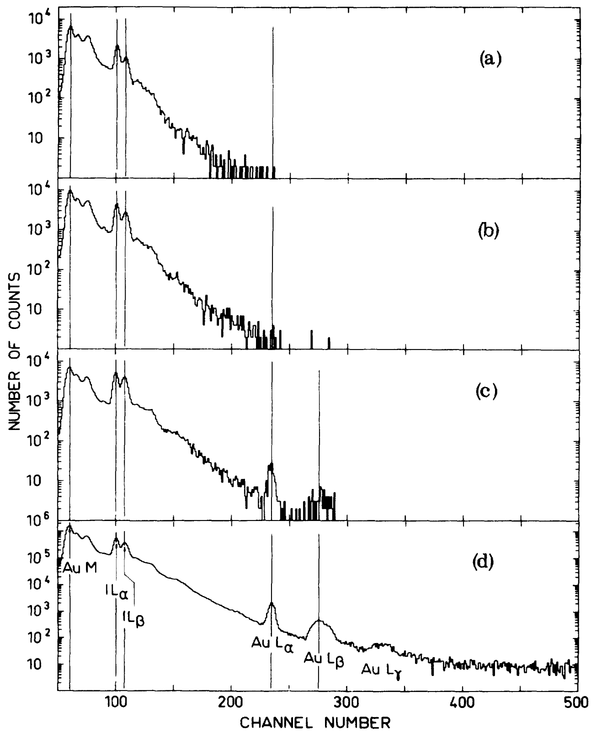

FIG. 1. X-ray spectra produced by bombardment of a 200-ug/cm^2 Au target with a beam of 26.85-MeV iodine. (a), (b), and (c): x rays in coincidence with iodine ions scattered at laboratory angles of 1.50, 3.52 and 10.50 deg., respectively; (d): singles spectrum.

We have proceeded in the following way: Multiple-scattering effects have been calculated by use of the formalism developed by Meyer.[4] For a two-step reaction mechanism we approximate the production cross section for the I L vacancy required in this model[1] by the use of a screened Coulomb potential[4] and the L-vacancy probabilities measured by Stein *et al.* for the case of I+Te.[5] This part of the calculation is not necessary for a one-step process. The cross section for production of the MO x ray is taken to be given by a shielded Coulomb cross section and a production probability which is essentially constant in the angular range of interest. For a given scattering angle a simple numerical integration then gives the spectrum of particles producing MO x rays as a function of impact parameter. It is found in all cases that there is a well defined cutoff on the minimum impact parameter and that this value is close to that calculated from the actual scattering angle. We feel that the theory of MO x rays is not yet detailed enough to permit the calculation of a detailed fit to the coincidence line shape. It is assumed that the energy of the MO x ray at the minimum impact parameter corresponds to the endpoint energy and is not shifted appreciably by collision broadening.[6,7]

III. DISCUSSION AND CONCLUSIONS

The spectra shown in Figs. 1 and 2 confirm the interpretation of the x-ray singles spectra dis-

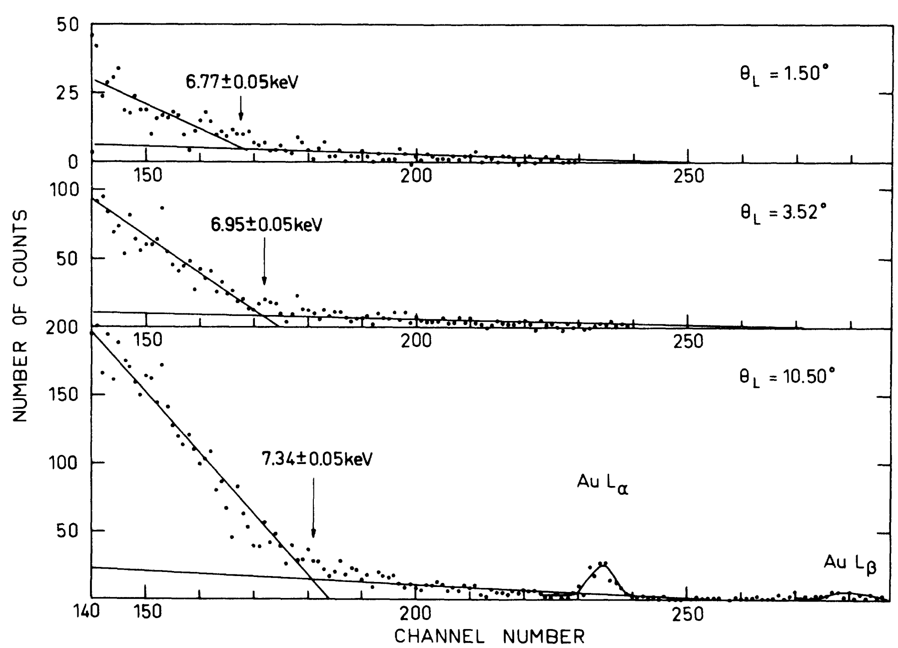

FIG. 2. Spectra showing magnified view of coincidence spectra for energies above 5.9 keV. The angles of the scattered particles are shown. Other features of the spectra are discussed in the text.

cussed by Mokler and co-workers[1,2] and Jundt *et al.*[3] A band of x rays is clearly observed with endpoint energies which increase with decreasing distance of closest approach, as would be expected for MO x rays. A value between 7.5 and 8.0 keV is found, by a small extrapolation, for the energy at a distance of closest approach small compared to the united-atom M-shell radius and should therefore be close to the united-atom M-x-ray energy. This energy agrees well with the centroid energy of the x-ray band observed in the singles experiments,[1,2] but can be determined in the coincidence measurement more precisely and unambiguously. The variation of the endpoint energy with distance of closest approach, gives directly the radial dependence of the molecular orbitals which produce the $4f$ - $3d$ transition in the quasiatom. The experimental observation that the endpoint energy does not vary strongly for small values of impact parameter is in qualitative agreement with the theoretical calculation of Fricke *et al.*[8]

The position of the endpoint was determined by a linear least-squares fit to the data with the assumption of a linearly decreasing background for the energies above 8 keV. The slight toe which is observed in all the coincidence spectra is attributed to the effect of the finite Si(Li) x-ray detector energy resolution.

At all three angles we observe a slowly decreasing background above the endpoint of the molecular orbital. The possibility that this background arises from improper subtraction of accidentals has been considered and is thought unlikely. It is possible that it results, at least in part, from a geometrical summing of an iodine L x ray with a MO x ray produced by the same iodine atom. The statistics are too poor to give an accurate determination of endpoint in this case. An alternative explanation could possibly involve in part the effects of collision broadening as discussed for the MO x rays produced in Ni-Ni collisions.[6,7]

An estimate of the energy to be expected for the M x ray in element 132 has been made with a Hartree-Fock-Slater atomic configuration program which includes relativistic and spin-orbit corrections.[9] The starting potential was determined by extrapolation from element 102. At a bombarding energy of 26.85 MeV, the most probable charge state of iodine is 14.[10] If it is assumed that there is one vacancy in the $3d$ shell, then an x-ray energy of 7.331 keV is found for the $4f$-$3d$ transition. When there are two vacancies in the $3d$ shell and a resulting charge of +15, the energy of the $4f$-$3d$ transition increases to 7.844 keV. Thus the theoretical calculations agree well with the experimental estimate, but show that the experimental value can depend sensitively on the exact electronic configuration of the transient molecule. This conclusion has been reached previously by Fricke and Waber.[11]

The measurements at three different angles constitute a crude angular distribution and we are able to find the total cross section for MO x-ray production from our measured value of probability for producing the MO x ray, P_x, by the equation $\sigma_x = 2\pi \int_0^\infty P_x\, b\, db$, where b is the impact parameter. Our data give a very approximate value of 750 b which is in reasonable agreement with the values of 100 and 160 b found in the singles experiments.[1,2]

Our data also show that there is a strong angular dependence for the probability of producing Au L x rays. This agrees with the size of the Au L-shell radius and the distances involved. We find no evidence here for the formation of L or K x rays which could be attributed to the quasiatom with $Z = 132$. A limit for the cross sections is put at 10^{-2} of the cross section for production of the quasiatom M x ray.

In this work we have observed for the first time MO x rays in coincidence with the scattered ions which produce them. The addition of the coincidence requirement makes possible the acquisition of more precise and detailed data than is possible with a singles measurement and opens new possibilities for precision spectroscopy of M x rays of superheavy quasiatoms. The feasibility of making similar measurements on L and K MO x rays appears to be less because of the much reduced cross sections for these processes.

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