MASTER

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

UCRL - 73823
PREPRINT

CONF-720404-8

LAWRENCE LIVERMORE LABORATORY
University of California/Livermore, California

Production of Characteristic X Rays by Heavy Ion Bombardment

T. M. Kavanagh

R. J. Fortner

R. C. Der

March 22, 1972

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

This paper was prepared for presentation at
International Conference on Inner Shell Ionization Phenomena
April 17-21, 1972, Atlanta, Georgia

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Production of Characteristic X Rays by Heavy Ion Bombardment*

T. M. Kavanagh, R. J. Fortner, and R. C. Der

Lawrence Livermore Laboratory, Livermore, Calif. 94550

Abstract

Data on inner-shell vacancy production in heavy ion-atom collisions, as obtained from x-ray measurements, will be reviewed. Cross section data and results of spectral measurements will be discussed and related to a parameter $E/\lambda u$ that indicates the degree of adiabaticity of the interaction. Interaction mechanisms will be examined in this context.

*Work performed under the auspices of the U. S. Atomic Energy Commission.

In this paper we will review some of the contributions that have been made by x-ray studies to our understanding of inner-shell ionization in heavy ion-atom collisions. Several features of this interesting class of phenomena have been discussed in preceding papers in this conference, and other papers on the subject follow this one. An attempt will be made here to bring together various aspects of the subject that have been studied in our Laboratory (though data from many laboratories will be discussed) and to give a view of what we feel has been learned, and in which directions we feel further work should be channeled.

Inner-shell excitation by the lightest ions, protons and alpha particles, is relatively well understood, and will be discussed only briefly here. A discussion of the proton case has been presented in an earlier paper.¹ In general the mechanism is viewed as a direct Coulomb interaction between the incident ion, assumed to be a point charge, and the bound electron, and it has been quite successfully treated in Born approximation and more recently in the binary encounter theory of Garcia.² In Fig. 1 we show the experimental K-shell data³ for incident protons, plotted using Garcia's scaled parameters (E and u are the proton and K electron energies, respectively, λ is the ratio of the proton to electron mass and Z is the projectile charge); the experimental data collapse extremely well onto the binary encounter curve, which is shown as a solid line in the Figure.

The situation is quite different for incident heavy ions. Insofar as x-ray measurements are concerned inner-shell vacancy creation differs from the proton and alpha particles case in two fundamental ways:

1. Cross sections are in general much larger for the heavy-ion

case—approaching geometric values for incident ion velocities that are much smaller than the relevant electron velocities. The cross sections show very strong fluctuations with Z values of the collision partners, with maxima occurring when the binding energy of the electron being excited matches the binding energy of some electron in the collision partner.

2. There is a much higher probability, in the heavy-ion case, of producing multiple vacancies.

We illustrate the situation regarding total cross sections for heavy-ion bombardment in Fig. 2, in which we have plotted most of the K-shell ionization cross sections that have been obtained from x-ray measurements.³ The data are plotted in terms of the reduced variables used by Garcia in his binary encounter theory (see Fig. 1). This kind of plot is used here for two reasons: First, it shows the disagreement of the heavy-ion data with a model that is very successful for incident point charges. Secondly, this plot brings together data extending over an extremely broad range of bombarding energy and K-electron binding energy and displays them in terms of the parameter $E/\lambda u$ which quite directly indicates the degree to which the interaction can be considered adiabatic (i.e. $E/\lambda u \equiv (v_0/v)^2$, where v_0 and v are the ion and electron velocities, respectively). The assumption of quasi-adiabaticity is, of course, the essence of the molecular-orbitals, electron-promotion model⁴ commonly used in the interpretation of heavy ion data; the plot thus shows directly the range of applicability of this point of view.

The solid curve in Fig. 2 represents the predictions of the binary encounter theory. We have shown in Fig. 1 that this theory agrees extremely well with experimental K-shell data for incident protons; clearly this is not the case for heavy ions. The majority of the heavy-ion data fall into a broad band that exhibits a flatter energy dependence than the theory, and that is several orders of magnitude higher in cross section. At the higher energies the data appear to merge with the theoretical curve, and there is a rapid rise at lower energies. (The more correct use of a screened nuclear charge, instead of Z , in the vertical scale parameter would increase this disparity between the theory and experiment). In addition to this broad band of data we show, in Fig. 2, data for oxygen ions incident on Al, Ca and Cu; note that this plot appear to collapse all these oxygen ion data into a narrow band, much different than that for the other heavy ions, and more closely resembling the theory for incident point charges. It is worth noting that a correction of the oxygen data for the adiabatic effects discussed by Brandt, Laubert and Sellin⁵--the so-called "binding correction"--should bring them into better agreement with the theory; the same correction would, however, increase the disparity for the other heavy-ion data in Fig. 2.

In the molecular-orbitals, electron-promotion scheme the large cross sections for heavy ion-atom collisions are assumed to result from transitions at crossings of molecular orbitals; examples of such level schemes will be presented later. Implicit in this formalism is the requirement of small $E/\lambda u$ --i.e. the requirement that the bound electron can adjust nearly adiabatically to the time-varying potential that it experiences due to the proximity of the other nucleus. Note that the bulk of the data in Fig. 2 are for $E/\lambda u \lesssim 0.1$, and for $E/\lambda u \lesssim 0.01$ the molecular orbitals picture can be expected to have a high degree of validity .

(For $E/\lambda u < 0.01$ the relevant electron would complete 10 or more orbits in its unperturbed configuration during the collision.) If deviation of the heavy ion data from the binary encounter predictions is taken as a guide, Fig. 2 perhaps indicates the importance of molecular orbitals effects for $E/\lambda u$ as large as 0.1. While the data in Fig. 2 represent experiments performed with bombarding energies from a few keV to several tens of MeV, there are few data for $E/\lambda u > 0.1$ and none for $E/\lambda u > 1.0$; it is in this region that the molecular orbitals picture is invalid and it is interesting that the experimental data appear to merge with the binary encounter curve. Further, higher energy data should be collected.

The data in the broad band in Fig. 2 can be collapsed onto a single theoretical curve based on a very simple level-crossing model that uses the Landau-Zener formulation for the transition probability at a crossing.⁶ This fit is shown, for carbon K-shell data, in Fig. 3; equally good fits can be obtained for most of the data in the "broad band" in Fig. 2. The agreement, excellent for $E/\lambda u$ as large as 0.1, is somewhat surprising in view of the simplicity of the Landau-Zener theory (a two-level theory, assuming a single level crossing); the success of this parameterization may, however, contain implications as to the required contents of a more rigorous theory.

In Fig. 4 we show a curve like that in Fig. 2 but for L- and M-shell data.³ The deviation from point charge, direct Coulomb interaction theory is perhaps even more marked here. These data, which extend to smaller $E/\lambda u$ values than in the K-shell case more clearly show the rapid rise for small relative velocities. The Landau-Zener theory that was successful in the K-shell case does not fit these data; this can perhaps be

attributed to the fact that the assumption of a single level crossing is not valid in these cases.

The reasons why the oxygen data shown in Fig. 2 appear to agree with predictions for incident point charges, whereas the other heavy ion data do not, is easily understood in terms of the phenomenon of "level matching."⁷ This effect is illustrated by Fig. 5 which shows the cross section for the production of L x rays in 160 keV copper ions incident on a broad range of targets. Prominent maxima occur for target Z-values such that the copper L-shell binding energy matches with target L- and M-shell energies. (These matchings are indicated by the dashed lines in the Figure, which show binding energies, in accordance with right-hand scale; the horizontal dashed lines represent copper L-shell energies and the dashed curves are target binding energies as a function of Z_2 .) Other data show a similar peak for K-L matching^{7,8} and cross sections are large in the region of K-K matching (see the C^+-C data in Fig. 2).

The significance of the locations of level-matching peaks is clearly seen from an examination of molecular orbitals diagrams for asymmetric collisions; we show a set of such diagrams, relevant to L-shell excitation in argon, in Fig. 6. (These are plots of level energies as a function of inter-nuclear distance r_0 , with very small distance on the left, and very large distance on the right. Barat and Lichten⁹ have recently discussed the rules for constructing these diagrams for asymmetric collisions. Separated-atom levels correlate with united-atom levels that have equal numbers of nodes in their radial wave functions.) Large cross sections are assumed to result for a given electronic excitation when that electron is "promoted" along a steeply rising molecular orbital that crosses with other infilled levels. Considering the creation of 2p vacancies in argon,

in going from Fig. 6(a) to Fig. 6(b) we pass through the K-L match; in Fig. 6(b) we see that argon 2p electrons are promoted along the 3d σ orbital, whereas on the low-Z side of the level matching--Fig. 6(a)--it is the boron 1s electrons that are promoted via the 3d σ . Figs. 6(c) and 6(d) span the L-L match which involves a switch of argon 2p electrons onto the very steeply rising 4f σ orbital. The situation regarding the K-shell excitation by incident oxygen ions, discussed in connection with Fig. 2, can be seen, by analogy, with the behavior of the argon K shell in Figs. 6(a), (b), and (c). The argon 1s level is the most tightly bound level in the system, not near a level matching, and no promotions or level crossings occur. Adiabatic effects simply cause an increase in the electron binding energy, and any excitation is expected to occur by mechanisms like those operative for incident protons. (This binding energy increase is the effect requiring the "binding correction" referred to earlier.)

We emphasize that while diagrams like those in Fig. 6 show switches in level ordering, with resultant changes in promotion schemes, corresponding to locations of level matching peaks in the cross-section data, a direct application of these concepts would predict an abrupt rise in cross section at a level matching. There is, however, an onset of the effect prior to precise matching, with a width much too large to be explained by a simple application of the uncertainty principle.

As pointed out at the beginning of this paper, a further interesting feature of the heavy ion x-ray data is the high probability, in these collisions, of creating multiple vacancies. These effects have already been discussed in preceding papers,¹⁰ and there is extensive evidence

in the literature which shows that while proton bombardment of a given target produces a single characteristic x-ray line much like that produced by electron or photon excitation, heavy ion bombardment results in shifted and broadened lines, and in the creation of totally new lines. X-ray components with energies in excess of the normal ground state binding energies for a given shell are observed. The reasons for the shifts are, of course, that the removal of outer-shell electrons changes the degree to which inner-shell electrons are screened from the nuclear charge. Hartree-Fock type calculations of energies for excited configurations have been carried out quite extensively, and a great deal of detailed information has been accumulated regarding complex defect configurations. Rather than discuss x-ray spectra and their interpretation in terms of defect configurations (such things are the subject of the papers in a preceeding session¹⁰), we wish to simply make a few remarks in an attempt to relate these data to our understanding of the mechanisms involved in the inner-shell vacancy creation.

Some of the x-ray spectroscopic data give strong support to the molecular orbitals interpretation. Low energy collisions involving argon¹¹ show multiple excitations in which an L vacancy is produced, and an electron is placed in the normally empty 3d level. The resulting spectrum is illustrated in Fig. 7 which shows the spectra of L x rays from Ar^+-Ar collisions over a range of ion energies. The lower energy peak, most prominent in the spectrum for 50 keV bombardment, is the usual $L_{2,\eta}$ line (3s $\rightarrow$ 2p transition) and, on the basis of Hartree-Fock-Slater calculations, the higher energy line is designated¹¹ as the $L_{\alpha,\beta}$ components (3d $\rightarrow$ 2p)

normally not observed in argon. Other x-ray components appearing in the spectra for higher bombarding energies are interpreted in terms of multiple L- and M-shell excitations. Note that the $L_{\alpha,\beta}$ component, that appears in all the spectra, has an energy in excess of the ground state 2p binding energy of argon. The excitation of an electron into a higher level as implied by this component (i.e. as opposed to excitation into the continuum) is, of course, a natural consequence of a molecular orbitals promotion scheme.

While the molecular-orbitals, level-crossing mechanism can, in principle, provide multiple excitations in a straight forward way (since many crossings of many levels are involved in the quasi-molecular interaction) care must be exercised to avoid using this concept outside the range of its validity. In most of the data in the literature, the creation of the primary vacancy (i.e. the one that produces the x ray being examined) can perhaps be considered quasi-adiabatic (i.e. $E/\lambda u \lesssim 0.1$) but such is not always the case for the outer-shell vacancies that are apparently created at the same time and that are responsible for the observed changes in x ray lines. For example, for 10 to 40 MeV oxygen ions incident on Cu, $E/\lambda u$ is ~ 0.1 for the K shell, but for the L and M shells that are excited simultaneous with K-shell excitation, the $E/\lambda u$ values are of the order of 1 and 10 respectively. Fig. 8 shows spectra of L x rays of Cu, Ni, and Zn produced by 15 MeV oxygen ions, with spectra for incident protons shown for comparison; analysis of the data¹² indicates creation of double L-shell vacancies and as many as 12 M and N shell vacancies. It is not clear at this time why the observed multiple excitation probabilities for such collision systems should be so much larger than for incident protons. (Recall that insofar as K-shell excitation cross sections are concerned for such a

collision system, the incident oxygen ion is apparently like a point charge--see Fig. 2.) It is in this area that we see a special need for theoretical insight. Why should an oxygen ion in the multi-MeV energy range behave like a point charge insofar as its interaction with the copper K shell is concerned, while at the same time exhibiting a much larger propensity than a proton toward simultaneous disruption of the outer shells? It is clear that the explanation lies in the interaction of the bound electrons of the two collision partners, but there is at present no theoretical framework useful in describing such effects for non-adiabatic situations.

References

1. E. Merzbacher, Conference proceedings.
2. J. D. Garcia, Phys. Rev. A1, 280 (1970).
3. R. C. Der, R. J. Fortner, T. M. Kavanagh, J. M. Khan, and J. D. Garcia
Rev. Mod. Phys. (to be published).
4. U. Fano and W. Lichten, Phys. Rev. Letters 14, 627 (1965).
5. W. Brandt, R. Laubert and I. Sellin, Phys. Rev. 151, 56 (1966).
6. R. C. Der et al., Phys. Rev. Letters 24, 1272 (1970).
7. T. M. Kavanagh et al., Phys. Rev. Letters 25, 1473 (1970).
8. F. W. Saris, Physica 52, 290 (1971).
9. M. Barat and W. Lichten, Phys. Rev. (to be published).
10. P. Richard, Conference Proceedings
D. J. Nagel, Conference Proceedings
11. M. E. Cunningham et al. Phys. Rev. Letters 24, 931 (1970).
12. R. C. Der et al., Phys. Letters 36A, 239 (1971).

Figure Captions

Fig. 1 Comparison of experimental K-shell ionization cross sections for incident protons with the binary encounter theory of Garcia.² E and ϵ_K are, respectively, the proton energy and the K-shell binding energy, λ is the ratio of the projectile to electron mass and Z is the projectile atomic number. All the available data³ are included, and the theory is represented by the solid curve.

Fig. 2 Cross sections for K-shell ionization by incident heavy ions.³ The scaling parameters and the theoretical curve are the same as in Fig. 1. All data are based on x-ray measurements except that for Na^+-Na , which includes some Auger-electron data.

Fig. 3 Comparison of K-shell excitations in carbon with predictions of a simple level crossing model based on the Landau-Zener theory (from Ref. 6). The parameter v is the ion velocity, y is a parameter that depends on the dynamics of the specific level crossing, r_x is the level crossing radius and α is a statistical factor.

Fig. 4 Cross section data for L and M shells³ plotted as in Fig. 2. All data are from x-ray measurements except that for Ar^+-Ar which comes from Auger electron studies.

Fig. 5 Cross sections for copper L x-ray production (left-hand scale) in incident copper ions striking metal targets, as a function of the atomic number of the target, for a fixed ion energy of 160 keV (2.5 keV/amu). The dashed lines represent ground-state electron binding energies (right-hand scale): The horizontal dashed lines represent copper L-shell binding energies, and the dashed curves show target atom binding energies as a function of target atomic number (from Ref. 7).

Fig. 6 Molecular orbitals diagrams for collisions of argon with (a) boron, (b) carbon, (c) aluminum, and (d) calcium.

Fig. 7 Spectra of L x rays produced in argon-argon collisions, for several different bombarding energies (from Ref. 11).

Fig. 8 L x rays produced by 15 MeV oxygen ions incident on nickel, copper and zinc targets. In each case the L x rays produced by 1.6 MeV protons are also shown (from Ref. 12).

Fig. 1

Fig. 2

Fig. 3

Fig. 5

Fig. 6

Fig. 7

Fig. 8