Characteristic x rays due to projectiles moving through solids

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(Received 19 January 1979)

The assumptions used in a previous study on the analysis of characteristic x rays emitted by a projectile while traversing a solid are reexamined. New information, in addition to accompanying comments by another author, is considered. While more work is needed on the subject, it is concluded that these new studies support the potential use of characteristic x rays as diagnostic tools for the study of ions traveling through solids.

In a previous paper[1] we attempted to show that x rays resulting from inner-shell vacancy filling could be used as a tool for investigating some aspects of excitation states of argon ions while they are traversing a carbon solid. As we stated then, the problem is quite a complex one, and several assumptions were necessary to make any analysis possible. We tried to make explicit the various assumptions being made, and we have pursued subsequent research efforts aimed at testing some of these assumptions. In this note, we comment on a few aspects of this research. We still are of the opinion that study of these x rays provides useful information concerning projectile excitation states.

The primary assumption (designated A1) in our analysis was that the dominant physical phenomena associated with inner-shell excitation could be separated from those attendant the outer-shell electrons. This enabled us to consider the inner-shell events as occuring in sequential binary collisions between the projectile and a target atom, and permitted us to write rate equations, our Eqs. (1) and (2), for occupation numbers for the projectile inner shell. These equations were predicated on the fact[2] that in gas-target collisions there is no evidence for carbon K-shell x rays in Ar-C collisions. This can be understood in terms of the molecular-orbital-promotion model [see Fig. 4(a) in Ref. 1], which predicts that Ar $2p$ electrons are promoted via the $3d\sigma$ molecular orbital, while the carbon $1s$ electrons are not promoted (A2). We made a subsidiary assumption (A3) that some of the Ar projectiles *not* having an inner-shell vacancy, but having a sufficient number of outer-shell vacancies, could cause carbon $1s$ electron promotion instead. This brought in the factor α in Eq. (4). We also assumed (A4) that Ar projectiles with one or more inner-shell vacancies would also promote the carbon $1s$ electron.

These assumptions, together with total cross-section measurements and stopping-power data, permitted us to obtain expressions for the target and projectile x-ray yields in terms of the unknown Ar dynamic flourescence yield $\overline{\omega}_p$, the total vacancy lifetime τ, and α. These also were assumed constant (A5), and a fit of the two thick-target yield curves yielded values for these parameters. A by-product of this effort was the fraction of ions having an inner-shell vacancy as a function of distance into the solid. An implicit assumption in the above, (A6), the neglect of carbon x rays due to recoil, was based on an analysis by Taulbjerg and Sigmund.[3]

In our thick-target-yield analysis, the x rays attributed to carbon K-shell emissions were separated by decomposition of the spectra. We attributed all of the x rays at the position of the carbon K-shell line to carbon emissions. Barragiola, in an accompanying Comment,[4] questions this decomposition. While it is true that the argon $3d$-$2p$ transition from the initial configurations $1s^2 2s^2 2p^5 3s^2 3p^n 3d$ contribute x rays in this spectral range, at least in gas targets[3] these transitions contribute only a small percentage of the total. Our carbon thick-target yields should, however, be considered an *upper* bound. As regards the line profile, the peak at the position of the carbon peak has a width consistent with the instrumental profile for that experiment, so any broadening would not have been observable in that experiment. (See below.)

Assumption (A1) forms the basis of most of the current quantitatively successful interpretations of inner-shell vacancy production in heavy-ion–atom collisions; i.e., it is the framework used to reduce the many-electron problem to a one-electron problem even for single-collision-region gas-target experiments. We consider it justified in this application as well. Assumption (A2) is borne out by gas-target data[2] for $Ar \rightarrow CH_4$ as well as $C^+ \rightarrow Ar$

collisions. These data indicate that the carbon 1*s* and argon 2*p* electrons retain their correlation identity, and refute the claim that the probability for each level to be promoted via the $3d\sigma$ orbital should be nearly the same, as claimed by Barragiola.[4]

It should be noted that assumption (A3) is in contradiction to assumption (A1). We were concerned about this, and have performed additional experiments in which carbon gas targets were used, and the charge of the Ar projectiles was varied.[5] The results of these experiments indicate that assumption (A3) is incorrect. No "outer-shell swapping" of the 2*p* Ar and 1*s* C levels was observed. [Again these data indicate that those levels do retain their correlation identity and only the Ar level is promoted, so that Barragiola's processes (i) and (iii) do have nearly the same cross sections.]

The fact that our analysis yielded a small, but nonzero, α is probably due to our assumption (A6) of neglecting recoil. We agree with Barragiola that recoil is probably important at our lowest energies and must be included in a proper analysis. (α was only important for the low-energy data, as stated in Ref. 1.) The rapid growth of the carbon peak versus projectile energy is, however, not attributable to recoil, as the ratio of direct to recoil yield should *decrease* with increasing energy. Assumption (A4) needs still to be separately experimentally tested, but we believe that, in accord with (A1), the inner-shell phenomena can be treated separately and that this mechanism is in fact the reason for the increase in the carbon yield. Assumption (A5), with respect to the constancy of τ needs substantial work. We agree with Barragiola that a three-level analysis including the possibility of vacancy sharing should be done, and incorporated into the analysis.

In our analysis of the x-ray spectra, we commented that the unresolved nature of the spectrum needed further investigation. We have investigated the nature of the spectrum for solid versus gas targets, using higher-resolution bent-crystal spectrometers and using projectiles whose lines in gas-target experiments are easily well resolved by the crystal spectrometer. The results of these studies have been published elsewhere.[6] No evidence was found for structure within the broad features associated with the solid-target spectra. Broadening mechanisms are being studied; no simple answers are evident. For example, simple "immersion in the C valence band"[4] would depend on projectile velocity, and the valence-band structure might be observable under some circumstances. It is not evident that this is true. Much more work is needed on this topic.

Our spectral analysis led to a discussion of static versus dynamic flourescence yields as described by Eqs. (17) and (20), respectively, of Ref. 1. These were based on the fact that collisions could mix the initial states within a multiplet. This aspect was also investigated further,[7] using fluorine ions. In that experiment, it was found that the longer-lived metastable states were increasingly quenched, i.e., collisionally depopulated, relative to the short-lived states, as gas pressure was increased. This supports the concepts presented in Ref. 1. However, more studies are needed here also.

Barragiola, Ziem, and Stolterfoht[8] have used Auger electrons to arrive at a value for the number of Ar projectiles having an inner-shell vacancy after traversing a certain thickness of (solid) carbon foil. Their results are within a factor of 2 of the results predicted in Ref. 1. We consider these results to be in agreement, given the uncertainties in the analysis. As we stated in Ref. 1, our extraction of the argon *M*-shell populations was very approximate. It is not clear at this time, without extensive theoretical and experimental work, whether such a determination can be made as numerically precise as that for the *L*-shell. It should be noted, however, that charge-state populations extracted from the neon *K*-shell x rays produced qualitatively similar results,[6] i.e., the higher-charge states were also emphasized in that case.

It is evident that much more work is required in order to establish these techniques as quantitative working tools for studying states of ions while traversing a solid. However, given the fact that two independent methods produce results as close as a factor of 2, at least for the *L* shell, we are optimistic concerning future efforts at quantifying at least some aspects of this problem

[1] R. J. Fortner and J. D. Garcia, Phys. Rev. A 12, 856 (1975).
[2] R. J. Fortner, Phys. Rev. A 10, 2218 (1974).
[3] K. Taulbjerg and P. Sigmund, Phys. Rev. A 5, 1285 (1972).
[4] R. A. Barragiola, preceding comment, Phys. Rev. A 19, 2470 (1979).
[5] R. J. Fortner, F. Saris, and P. Woerlee (unpublished).
[6] R. J. Fortner, D. L. Mathews, J. D. Garcia, and H. Oona, Phys. Rev. A 14, 1020 (1976).
[7] R. J. Fortner and D. L. Mathews, Phys. Rev. A 16, 1441 (1977).
[8] R. A. Barragiola, P. Ziem, and N. Stolterfoht, J. Phys. B 9, 1447 (1976).