Broadening of projectile x-ray lines in solid targets*

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In this paper high-resolution K x-ray spectra for boron (1 MeV) and neon (90 keV) projectiles moving in solid targets are reported. The x-ray transitions from the moving ions are found to be substantially broadened, compared to gas-target spectral lines. Estimates of the extent of broadening are presented, and physical mechanisms responsible for it are discussed.

INTRODUCTION

In this paper we report comparisons of projectile K x-ray spectra for gas and solid targets, observed using crystal spectrometers. We have chosen projectiles whose gas-target x-ray spectra are relatively simple in that the structure in the x-ray spectra is clearly resolved and readily identified. The spectra obtained from projectiles moving in solid targets are found to be qualitatively different. These x rays are found to exhibit little or no structure, indicating substantial broadening of the x-ray lines. Estimates of the extent of the broadening are obtained, and a discussion of possible causes of the broadening is presented.

EXPERIMENTAL PROCEDURE

The details of the experimental procedure have been discussed elsewhere.[1,2] The boron K x-ray spectra were obtained using the University of Arizona's 2-MV Van de Graaff accelerator, and the neon K x-ray spectra were obtained using the Lawrence Livermore Laboratory's 100-keV dc accelerator. In both cases the target consisted of a gas cell maintained at a pressure of 5×10^{-2} Torr or a thick solid, as indicated. The x rays were analyzed using bent-crystal x-ray spectrometers with either a lead stearate (boron) or mica (neon) as the diffraction element. The detectors were flow-mode proportional counters with thin (2000 Å) windows. The instrumental full widths at half-maximum were 1.5 and 2.8 eV, respectively.

The resultant x-ray spectra were found to be extremely sensitive to the amount of incident beam used to obtain the spectra for a wide variety of solid targets. Generally, x-ray peaks associated with the projectile were found to grow in intensity as the amount of beam increased. These peaks were attributed to x-rays from implanted projectiles produced in subsequent symmetric collisions. (A previous high-resolution study,[3] using aluminum projectiles, also showed the buildup effects). The data for the three targets (diamond, titanium, and copper) discussed in this paper did not exhibit these effects, at least for the beam currents and data accumulation times used. Therefore only these spectra were selected for presentation.

EXPERIMENTAL DATA

In Fig. 1, the neon K x-ray spectra for three types of collisions, i.e., 1.3-keV electrons incident on a neon gas target and 90-keV neon ions incident on a neon gas target and a solid copper target, are shown. The gas target spectra exhibit narrow, well-resolved peaks, and the origins of these peaks have been discussed extensively in the literature.[4,5] Basically, the lines correspond to an initial state involving one K-shell and varying numbers of L-shell vacancies. The peaks are designated as such in Fig. 1, where KL^n corresponds to the x-ray transition from an initial state having one K-shell vacancy and n L-shell vacancies. (Multiplet structure of the vacancy configurations does provide components from one configuration under adjacent peaks,[6] but this effect is small.) However, these well-resolved discrete peaks are not observed for the neon K x-ray spectra emanating from ions moving in solid copper targets. The peak of this spectrum occurs in the region of the normal KL^0 peak; however, the peak extends in energy over a very broad energy range with essentially no structure detectable in the spectra. This spectrum is skewed toward the high-energy side, probably indicating the existence of states of excitation higher than KL^0. On the low-energy side of KL^0 the spectrum extends to lower energies, where no gas target components are evident. This indicates that the individual lines are considerably broadened. A more detailed

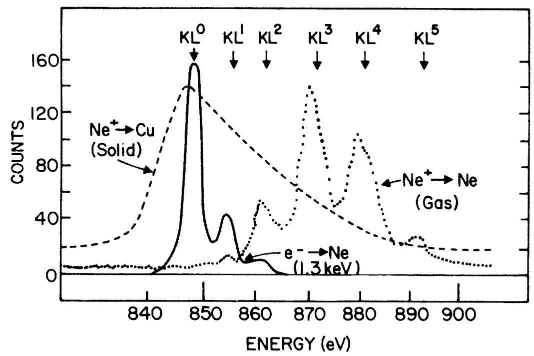

FIG. 1. Comparison of neon x-ray spectra produced by electron bombardment of neon gas (solid line), 90-keV neon ions on neon gas targets (dotted curve), and 90-keV neon-ion bombardment of solid copper targets (dashed curve).

discussion of the analysis of the spectra is presented below.

In Fig. 2 the 1-MeV boron K x-ray spectra for two types of boron-carbon collisions are presented. The first spectrum is for a methane gas target. An analysis of this spectrum has been presented elsewhere.[2] The x-ray lines are primarily from one-electron transitions in one-, two-, and three-electron boron. The second spectrum is for a solid diamond target. The most obvious difference is the large carbon K line which dominates the solid target spectrum but is not observed in a gas target. This peak occurs in the solid targets because of recoiling carbon atoms and subsequent C-C collisions,[7] and also because of the "swapping" of the boron 1s and carbon 1s binding energies. The increased boron 1s binding changes the correlations of the molecular orbitals and provides a channel for direct carbon 1s promotion. However, the differences of interest for this paper are the x-ray transitions emanating from the boron projectile. For the solid target, two broad continuous x-ray bands in the region where boron K x-ray lines were expected are observed. The low-energy band, centered near 190 eV, is consistent with calculations of $2p \rightarrow 1s$ x-ray transition energies in atoms having initial states involving one K-shell vacancy and one, two, or three electrons in the L shell. The higher-energy band, centered near 245 eV on the low-energy tail of the carbon K x-ray, is consistent with two possible identifications. The first involves transitions from highly excited states (i.e., $np \rightarrow 1s$ transitions, where $n \geq 3$) in atoms having one K-shell vacancy in the initial state. The second possibility involves $2p \rightarrow 1s$ transitions in atoms having two K-shell vacancies in the initial state and one, two, or three L-shell electrons. We believe that the x rays come predominantly from

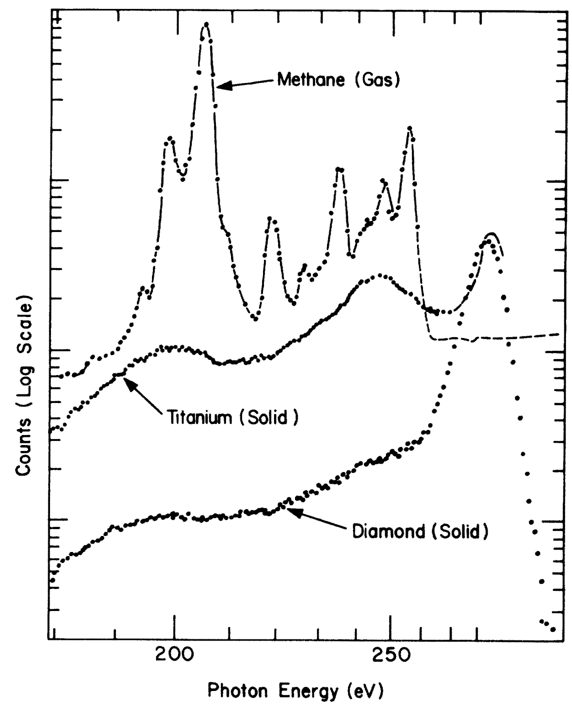

FIG. 2. Spectra produced by 1-MeV boron ions in methane gas, solid titanium targets, and solid diamond targets.

atoms having two K-shell vacancies. If the transitions were from single-vacancy, high-excitation states, the most prominent transitions would be $1s3p \rightarrow 1s^2$ (235.4 eV) and $1s2s3p \rightarrow 1s^2 2s$ (219.7 eV), as evidenced by the gas spectra. These energies do not agree well with centroid energy of the band. On the other hand, double K-shell vacancies transition energies are K^2L^2 (255.2 eV), K^2L^1 (~247.8 eV), and K^2L^0 (~242.6 eV). (Here K^2L^n refers to 2 K-shell, n L-shell vacancies.) These energies involving double K-shell vacancies agree well with the energy of the second band. Thus we tentatively identify the second band as x-ray transitions associated with double K-shell vacancies. The third spectrum in Fig. 2 was obtained using a solid titanium target. The spectrum still exhibits a carbon K x-ray peak which is due to carbon buildup on the target during the time the data were taken. However, the carbon K-shell peak is much smaller in this case. This spectrum shows the same two x-ray bands as in the diamond target case. We should mention that the broadening observed here is also evident in the Al→Ni data of Ref. 3, though it was not the focus of that study.

DISCUSSION OF THE DATA

A significant point to be extracted from both Fig. 1 and Fig. 2 concerns the widths of the x-ray lines obtained from the projectiles moving in solid targets. In both figures, the data obtained from gas target spectra exhibit clearly resolved peaks. However, when the x-ray spectra are measured using the same instrumental resolution for ions moving in a solid target, all the sharply resolved x-ray structure disappears and only broad x-ray bands are resolved. This broadening of the x-rays in the solid is the x-ray analog of the broadening of spectral lines observed in dense plasmas.

An estimate of the size of the x-ray broadening can be obtained by unfolding the x-ray spectra in Figs. 1 and 2. In this unfolding of the data we used Gaussian line shapes and centroid energies as observed in gas target measurements and we assumed that the broadening for each line in a given spectrum was the same. This unfolding of the $Ne^+ \rightarrow Cu$ spectra indicated a width of 12 ± 3 eV. The boron K x-ray spectra were unfolded using the double K-shell vacancy peak from Fig. 2. The spectra indicated a width of 12 ± 5 eV and equal intensities of the K^2L^0 and K^2L^1 x-ray peaks. In the boron data the larger error is due to possible x rays involving excited states and K^2L^2 transitions which would increase the apparent widths. In both cases the instrumental contribution to the line width was much smaller than the observed width.

For the neon-copper data, we have used the relative intensities of the x rays at the positions of the KL^n x-ray lines (as observed in the gas target spectra), converted them to relative probabilities using appropriate fluorescence yields, and plotted them as a function of the number of L-shell vacancies. This is shown in Fig. 3, compared to the probabilities for equilibrium charge states of neon ions at the same velocity emerging from carbon foils. The fractions of projectiles in higher charge states is larger for neon ions in copper. This is similar to the result obtained for argon projectiles in carbon.[1]

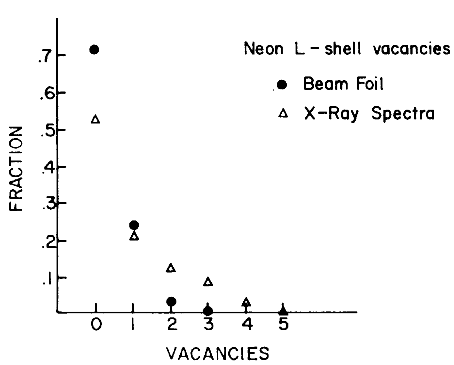

FIG. 3. Charge-state fractions for neon ions in solid copper targets and emerging from carbon foils. The foil data are from Ref. 11.

Another interesting aspect of the boron projectile data is the relative intensity of the single and double K-shell vacancy bands. For both the diamond and the titanium data the double K-shell vacancy peak is larger than the single K-shell vacancy peak. For gas target measurements the double K-shell vacancy lines are very weak. This indicates that the large enhancement of double K-shell vacancies in the solid is a multiple-collision phenomenon. Double K-shell vacancies can be produced in the projectile due to multiple collisions within the lifetime of an inner-shell hole. The atomic lifetime of a ground-state boron K-shell vacancy is 1.9×10^{-14} sec. This can be compared with the mean time for producing an inner-shell vacancy, $1/N\sigma v$, where N is the density of carbon atoms, σ is the K-shell vacancy production cross section, and v is the velocity of the projectile. The cross section is estimated to be 1.5×10^{-17} cm^2. This leads to a mean time for promotion of a K-shell electron in 1-MeV $B^+ \rightarrow C$ (diamond) collisions of $\sim 1.5 \times 10^{-15}$ sec. Thus double K-shell vacancies are quite probable, as the data show.

From the data, we find, after subtracting the carbon x rays, that the intensity ratio of double to single vacancies is 2.0 ± 0.4. If we assume that the outer-shell vacancy distributions for the two are the same, and if the fluorescence yields for single and double K-shell vacancy states in boron follow the same pattern as those in neon,[8] we estimate that the ratio of the fraction of projectiles having two K-shell vacancies to those having one is $f_2/f_1 = 1.9 \pm 0.4$.

BROADENING MECHANISMS

No detailed theoretical treatment of broadening mechanisms appropriate to projectiles moving in solids exists at this time. The primary difficulty lies in accurately assessing the role of many-body interactions, as opposed to binary collision mechanisms. In this section we estimate broadening due to the Doppler effect, the Stark effect, and to collisions with electrons and lattice ions. While some of these estimates must be considered unreliable, because of lack of a proper theoretical treatment, it appears that none of these mechanisms can account for the observed broadening.

It might be expected that Doppler broadening may be important in this situation, even though the spectrometers were at 90° with respect to the incident beams. The collisions which create the excitation states required for x-ray emission involve small impact parameters. Thus the fraction of projectiles being sampled by observing x rays may no longer be traveling in the original direction of the beam. An estimate of the broadening due to this effect can be made using conventional Doppler broadening theory:

$$\frac{\overline{\Delta E}}{E} \equiv \frac{\overline{\Delta \nu}}{\nu} = \frac{2}{\pi} \frac{\int_0^\pi \sin\theta\, P(\theta)[v(\theta)/c]\, d\theta}{\int_0^\pi P(\theta)\, d\theta}, \qquad (1)$$

where θ is the projectile deflection angle, c the velocity of light, $P(\theta)$ the probability of an emitter having an angle θ with respect to the beam, and $v(\theta)$ being the speed of the projectile moving at an angle relative to the beam. The factor $2/\pi$ comes from averaging over the azimuthal angle, assuming cylindrical symmetry. Because we are concerned only with those collisions which produce inner-shell excitations, the probability of an *emitter* being produced at small scattering angles is negligibly small. Thus we can replace the lower limit in this expression by θ_m, corresponding to projectiles for which K-shell excitation occurs with non-negligible probability. Furthermore, for such collisions the scattering angle is determined primarily by the Coulomb interaction between the nuclei. Thus a good approximation to the Doppler broadening is

$$\frac{\overline{\Delta E}}{E} = \frac{2}{\pi} \frac{\int_{\theta_m}^\pi \sin\theta\, P_c(\theta)[v(\theta)/c]\, d\theta}{\int_{\theta_m}^\pi P_c(\theta)\, d\theta}, \qquad (2)$$

where θ_m is the angle corresponding to interpenetration of the shell radii involved and $P_c(\theta)$ is the probability associated with Coulomb scattering of the nuclei. The angle θ here is, of course, the laboratory scattering angle. This expression provides an overestimate in that the probability of excitation corresponding to a given Coulomb deflection is set equal to unity and also because screening is neglected. Multiple scattering in hard collisions (assumed to be independent, random events) would increase the result by about 40%. The speed after collision, $v(\theta)$, is essentially independent of angle (except for nearly equal projectile masses, where it varies as $\cos\theta$). When the mass of the projectile, m_p, is much larger than that of the target, the upper limit in the integrals is to be replaced by $\sin^{-1}(m_p/m_t)$. For this latter case, therefore, Doppler broadening should be very small.

For $m_p \ll m_t$, we find

$$\frac{\overline{\Delta \nu}}{\nu} = \frac{4}{\pi} \frac{\cot\frac{1}{2}\theta_m + \frac{1}{2}\theta_m - \frac{1}{2}\pi}{\cot^2\frac{1}{2}\theta_m}. \qquad (3)$$

When $m_p = m_t$, this result should be reduced by a factor of 2.

The minimum angle θ_m is strongly velocity dependent; for example, for pure Coulomb scattering it is given by

$$\cot\tfrac{1}{2}\theta_m = mv^2 b/z_1 z_2 e^2,$$

where b is the impact parameter required for appropriate inner-shell penetration. Thus the result in Eq. (3) must be averaged over speed distributions appropriate to a projectile slowing down in the solid. If we assume that the stopping power is proportional to the speed (electronic stopping), and use this to determine the speed $v(x)$ as a function of distance into the solid, the leading (first) term in Eq. (3) leads to

$$\left\langle \frac{\overline{\Delta\nu}}{\nu} \right\rangle = \frac{\int_0^{x_{\rm th}} (\overline{\Delta\nu}/\nu)\, dx/v}{\int_0^{x_{\rm th}} dx/v} = \frac{\overline{\Delta\nu}}{\nu}\bigg|_{E_0} \left(\frac{(E_0/E_{\rm th})^{1/2} - 1}{\ln(E_0/E_{\rm th})^{1/2}} \right), \qquad (4)$$

where E_0 is the incident energy, $E_{\rm th}$ is the smallest energy which the projectile can have and still produce an excitation, and $x_{\rm th}$ is the distance into the solid at which $E_{\rm th}$ is reached. (A similar result can be obtained for the case that the stopping power is independent of velocity, a situation which sometimes occurs at low velocities when both nuclear and electronic contributions are important. It should be remarked that this expression (4) actually incorporates the salient features of the inner-shell excitation cross sections as well. These cross sections as functions of projectile energy generally rise steeply at some threshold energy, then exhibit only slight energy variation for larger projectile energies.[9]

The result of this analysis for the Ne-Cu data is that Doppler broadening should contribute no more than about 2 eV to the linewidth. Even smaller Doppler-broadening contributions are expected for the boron x rays.

Collisional broadening is also expected for the projectile x rays. However, estimates of this source of broadening are made difficult by the solid environment. Collisional broadening in plasmas has been extensively studied, and many of the relevant approximations for high-temperature, dense plasmas have been collected in Ref. 10. However, their applicability to the problem of an ion traversing a solid is questionable because of the binary collision basis used for many of them, and because the concept of beam temperature has little validity.

Some aspects of projectile-solid interactions can be handled, however, on a binary collision basis. An example of this is the excitation or ionization of a deep-lying level, where large energy transfer is required. For such a process the expected energy width can be written

$$\Delta E = \hbar\delta\nu = 2\hbar N\langle\sigma v\rangle, \qquad (5)$$

where N is the solid atomic density, σ the cross section, and where the brackets indicate a velocity average. We have applied Eq. (5) to the neon and boron ions, using $\sigma = \pi r^2$, where r is the mean radius of the upper state for the active electron. The resultant energy widths in both cases are of the order of 1 eV.

If we assume that the events responsible for the observed broadening are analyzable in terms of binary collisions, we can use Eq. (5) to find the averaged cross sections associated with such a broadening mechanism. For a 12 eV width, this results in

$$\sigma = \begin{cases} 3\times10^{-16}\ \text{cm}^2, & \text{for Ne-Cu}, \\ 1\times10^{-16}\ \text{cm}^2, & \text{for B-C}. \end{cases}$$

These cross sections are essentially gas kinetic cross sections, and are about an order of magnitude larger than the average cross-sectional areas of the upper levels for the states in question. This result indicates that binary collision mechanisms are not likely to be the dominant cause of the observed broadening.

Broadening due to collisions with electrons in the solid are much harder to estimate. Using the expressions of Ref. 10 with an electron temperature given by $kT_e = m_e E_i/m_i$ (subscripts e and i denote electron and projectile ions, respectively), we obtain widths of the order of 3–5 eV. However, these estimates cannot be considered reliable, and more theoretical effort is needed.

Stark broadening will also contribute to widths. Quadratic Stark effects (using plasma analysis) are negligible, and no linear Stark effect is expected for neon. For one- and two-electron boron, however, linear Stark effects may play a role. Using the expressions from Ref. 10 for the linear Stark effect due to lattice ions in the quasistatic limit, we find that this effect may produce a width of the order of 10 eV in the boron x rays. However, this estimate cannot be taken too seriously, because the boron projectiles are moving rapidly; thus a quasistatic limit is probably not appropriate. A better formulation of Stark broadening for projectiles moving in solids is needed.

Having examined the mechanisms which are normally responsible for line broadening, we find none which can be easily identified as being the dominant one for either the slow neon ions or the fast boron ions. The fact that Eq. (5) yields cross sections which are so large indicates that many-body interactions are likely to be important. Other mechanisms, such as charge exchange, may also play a role. It is evident that the current theoretical formulations are inadequate for this problem.

CONCLUSIONS

Our investigation of projectile x rays in solid targets, using high resolution, indicate that the spectra for neon and boron projectiles show broad features in the spectral region where characteristic x-ray lines for these projectiles are expected. The corresponding gas target spectra show well-resolved, identifiable x-ray lines. We have estimated the broadening of the lines, based on observed line spectra, and find the widths to be about 12 eV in both cases. Some of the mechanisms expected to be responsible for broadening have been examined. No single mechanism can at present be identified as being the dominant cause of the observed broadening. Further experimental and theoretical studies of this broadening could be helpful in clarifying the nature of the ion-solid interaction process.

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