Production of x rays beyond the K-series limit in gaseous and solid targets

J. Richard Mowat
Department of Physics, North Carolina State University, Raleigh, North Carolina 27650
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X rays of energy beyond the K-series limit have been detected during the bombardment of various low-Z, solid and gaseous targets by C^+, N^+, and O^+ ions of energy 0.75 to 1.9 MeV. $K\alpha\alpha$ x rays, most likely from the transition $(1s^{-2}2p^{-1}) \rightarrow (2s^{-1}2p^{-2})$, are emitted by *both* collision partners in gaseous targets. The low-resolution energy measurements are in good agreement with published calculations. The relative yield from the heavier collision partner is larger than simple vacancy-sharing models would indicate, but electron screening may account for the excess. Molecular-orbital (MO) continuum radiation completely obscures the $K\alpha\alpha$ lines from thin solid carbon targets. The exponential shape of the MO spectrum beyond the united-atom limit is in reasonable agreement with collision-broadened spontaneous emission.

I. INTRODUCTION

Intense interest in inner-shell atomic physics has been generated during the past decade, partly because of the need for x-ray spectroscopic data characterizing the highly ionized atoms commonly found in stellar and laboratory plasmas, and partly because of the emergence of the beam-foil technique as a method for producing highly ionized atoms without the use of high temperatures. High-velocity ion beams have been used to discover and study new features of the x-ray spectra of multiply ionized atomic systems. Many of these new discoveries have involved the observation of x rays, both discrete and continuous in energy, having wavelengths significantly shorter than, and in some cases very much shorter than, the wavelengths of the well-known characteristic x rays.

In 1975 Wölfli and co-workers[1] reported observations of faint new discrete-energy x rays emitted during collisions between swiftly moving Ni and Fe ions and stationary Ni and Fe target atoms. The x rays' energies were more than *twice* the characteristic $K\alpha$ x-ray energies of Ni and Fe. Those observations were interpreted as the first evidence for correlated, two-electron processes in inner-shell atomic physics. The authors suggested that a *single* high-energy x ray could be emitted when *two* L-shell electrons jump *simultaneously* into a *doubly* ionized K shell formed during the collision. The new x ray has been given the designation $K\alpha\alpha$.

Announcement of this discovery stimulated widespread theoretical and experimental research activity that confirmed the reality of correlated two-electron transitions within atomic inner shells. That work, concentrating on selected elements with atomic numbers between 7 and 28, has investigated the x-ray energies[2-9] and intensities[5-8,10] (relative to previously known x rays), the collision-induced x-ray yields,[11] and the quantum-mechanical character of the emission process.[2-5,7]

II. APPARATUS

The experimental arrangement is represented schematically in Fig. 1. A Si(Li) detector (resolution 210 eV at 6.4 keV) views either a commercial thin solid target (20 $\mu g/cm^2$ C or 20 $\mu g/cm^2$ Al on a 4-$\mu g/cm^2$ C backing) or a 400-mTorr windowless gas target (CO, N_2, O_2, or Ne) that is maintained by a capacitance-manometer-controlled servo valve. The target atomic numbers were chosen to bracket those of the projectiles. A single 12-μm Be window isolates the Si(Li) diode from the interaction region. The detector efficiency is estimated by using published mass absorption coefficients together with the manufacturer's estimates of the thicknesses and densities of the Be window, the Au contact, and the Si dead layer.

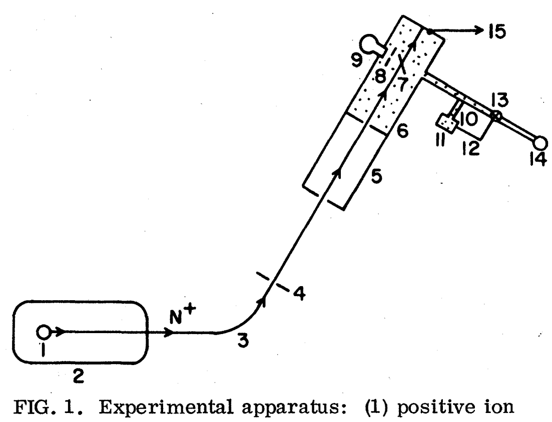

FIG. 1. Experimental apparatus: (1) positive ion source; (2) Van de Graaff accelerator; (3) analyzing magnet; (4) beam control slits; (5) high vacuum region ($p = 10^{-5}$ Torr); (6) gas target ($p = 4 \times 10^{-1}$ Torr); (7) thin solid target (20 $\mu g/cm^2$); (8) x-ray collimator; (9) Si(Li) detector; (10) gas sample; (11) capacitance manometer; (12) pressure control signal; (13) solenoid valve; (14) gas source bottle; (15) to beam current integrator.

The principal uncertainties in our experimental cross sections are due to imprecise knowledge of these thicknesses and densities. The detector energy calibration is based on spectra gathered during the bombardment of thick solid targets [F (i.e., Teflon), NaCl, MgO, and Al] with 0.5-MeV protons.

III. RESULTS AND DISCUSSION

A. Gaseous targets

Spectra taken from gaseous targets are dominated by peaks identified as $K\alpha\alpha$ x rays. Figure 2 is a raw spectrum obtained when 1-MeV N^+ ions pass through an N_2 target. The peak is identified by its energy[12] (893 ± 15 eV) as $NK\alpha\alpha$. The solid curve is a least-squares fit to a Gaussian plus a linear background. A weaker x ray of the same energy is produced when C^+ is the projectile so that N is the *heavier* collision partner. The Si(Li) detector window will not pass a detectable amount of $CK\alpha$, $NK\alpha$, $CK\alpha\alpha$, or projectile REC. In contrast to the spectrum published[13] by Hoogkamer *et al.* for 200 keV N^+ collisions with N_2, Fig. 2 displays no evidence for a MO continuum. The cross section for production of $NK\alpha\alpha$ by 200 keV N^+ bombardment of NH_3 has been reported[11] to be 1.4×10^{-25} cm^2. If the cross section scales as v^{-1} it should fall to 6.3×10^{-26} cm^2 at 1 MeV projectile energy. From the data shown in Fig. 2 we extract a cross section that lies in the range 2.2 to 6.1×10^{-26} cm^2, in reasonable agreement with expectations. The uncertainty is due to imperfect knowledge of the detector efficiency.

When 1.9 MeV N^+ ions pass through an O_2 target, a spectrum is obtained (Fig. 3) that contains $K\alpha\alpha$ peaks from *both* collision partners. No previously published work has reported the detection

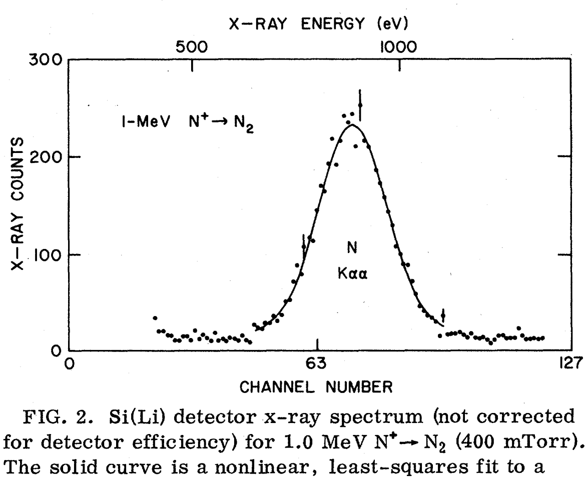

FIG. 2. Si(Li) detector x-ray spectrum (not corrected for detector efficiency) for 1.0 MeV $N^+ \to N_2$ (400 mTorr). The solid curve is a nonlinear, least-squares fit to a single Gaussian profile plus a linear background. Error bars represent counting statistics.

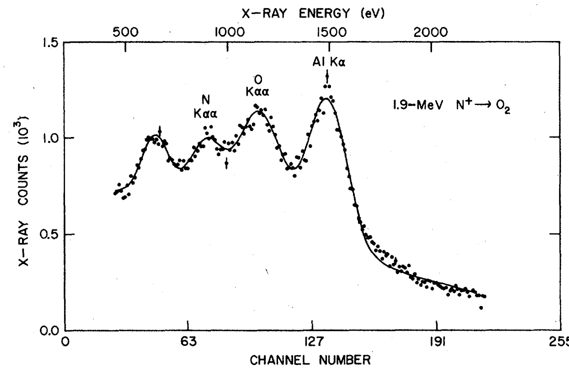

FIG. 3. Si(Li) detector x-ray spectrum (not corrected for detector efficiency) for 1.9 MeV $N^+ \to O_2$ (400 mTorr). The solid curve is a nonlinear, least-squares fit to the sum of four Gaussian profiles plus a linear background. Error bars represent counting statistics.

of $K\alpha\alpha$ x rays from the *heavier* collision partner when $Z<10$. The solid curve in Fig. 3 is a least-squares fit to the sum of four Gaussians plus a linear background. The measured $NK\alpha\alpha$ energy (893 ± 15 eV) and $OK\alpha\alpha$ energy (1160 ± 21 eV) agree well with the values computed by Hodge[12] (877 and 1145 eV, respectively) for the transition $(1s^{-2}2p^{-1}) \to (2s^{-1}2p^{-2})$. This assignment is reasonable since the incoming accelerated ion has a $2p$ vacancy prior to the collision that empties the K shell. The quoted experimental errors include both the scatter of fitted centroids from run to run and the uncertainty in the detector energy calibration.

The highest energy peak is $AlK\alpha$ and is produced when scattered projectiles strike the aluminum vacuum chamber walls. The lowest energy peak has not been identified, but its energy (652 ± 18 eV) corresponds to the $3p$-$1s$ transition $(1s2s3p)^2P^0 \to (1s^22s)^2S^e$ at 641 eV, which is an oxygen $K\beta$ satellite, or the $2p$-$1s$ transition $(2s^22p^2)^1D^e \to (1s2s^22p)^1P^0$ at 650 eV, which is an oxygen $K\alpha$ hypersatellite. Autoionization from these states has been observed by Bruch *et al.*[14] This peak is much more pronounced when the collision partners are interchanged. (Figure 4 is the spectrum obtained for 1.9 MeV O^+ bombardment of N_2.) The $NK\alpha\alpha$ and $OK\alpha\alpha$ relative intensities are unchanged which suggests that the rate of double K-shell ionization is the same as for N^+ bombardment of O_2. For this reason the identification of the 652 eV transition with the single K-vacancy state $(1s2s3p)$ is preferred. Figures 3 and 4 have not been corrected for the (x-ray energy dependent) detector efficiency. As discussed below, the $NK\alpha\alpha$ yield $Y(L)$ is actually greater than the $OK\alpha\alpha$ yield $Y(H)$.

Present models[15] of single K-vacancy sharing

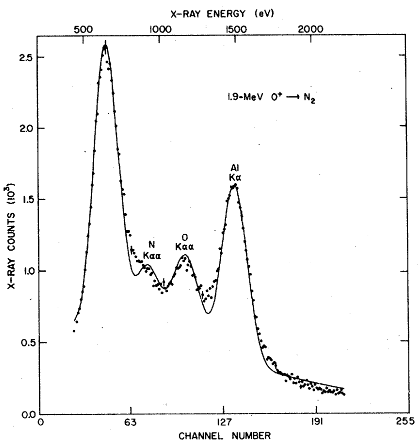

FIG. 4. Same as Fig. 3 except for the interchange of the target and projectile (i.e., $O^+ \rightarrow N_2$).

in ion-atom collisions, which have proved reliable for $Z_L + Z_H > 15$, place the majority of the separated atom K vacancies in the lighter collision partner. The ratio R_{KK} of the number of double K vacancies created in the heavier ion (H) to that in the lighter ion (L) is related to the experimental yields $Y(L)$ and $Y(H)$ by[16]

$$R_{KK} = \frac{Y(H)}{Y(L)} \frac{\omega_K^{0(L)}}{\omega_K^{0(H)}} ,$$

where the ω_K^0's are neutral atom fluorescence yields. Our value of R_{KK} based on the data shown in Figs. 3 and 4 lies between 0.2 and 0.4, where, again, the uncertainty is due to the imprecisely known detector efficiency. The Meyerhof model[15] as extended by Mitchell and Phillips[16] predicts $R_{KK} = 0.11$ for 1.9 MeV N^+ in an oxygen target. However, Boving[17] suggests correcting the Myerhof model to account for screening effects, when $Z_L + Z_H < 15$, by introducing the parameter $\sin\theta = \Delta U(R_{\min})/(I_H - I_L)$. In Meyerhof's model the true level spacing $\Delta U(R_{\min})$ at the distance $R_{\min}$ where $d(\Delta U)/dR$ passes through a minimum is approximated by $I_H - I_L$, the difference in separated atom K-shell binding energies. Our value of R_{KK} suggests that $\sin\theta$ lies between 0.95 and 0.98 for the system studied here ($Z_L + Z_H = 13$) compared to the Meyerhof value $\sin\theta = 1.00$. Boving finds $\sin\theta = 0.94$ and $\sin\theta = 0.97$ for $Z_L + Z_H = 11$ and $Z_L + Z_H = 18$, respectively.

B. Solid targets

The discovery[1] of $K\alpha\alpha$ x rays associated with the lighter collision partner occurred during the heavy-ion bombardment of solid targets with $Z \sim 26$. $K\alpha\alpha$ x rays from both collision partners were detected[18] during Cl bombardment of solid KCL while work[19] at lower Z (S and P bombardment of Si) found the $K\alpha\alpha$ lines to be obscured by MO continua. Another study[20] at still lower Z (O bombardment of Al) found no discrete features in the $K\alpha\alpha$ region.

All spectra [e.g., Fig. 5(a)] taken in the present experiments using 1 MeV C^+, N^+, and O^+ ions to bombard thin solid carbon targets are featureless continua (the detector window does not transmit detectable amounts of C, N, or O$K\alpha$). When carbon-backed aluminum targets are used [e.g., Fig. 5(c)] Al$K\alpha$ appears along with a lower-energy, featureless continuum which is entirely due to the carbon backing. The measured spectra do not extend to the region beyond the projectile-aluminum united-atom limit. Al$K\alpha$ cross sections have been extracted and found to be ~1 b. This value is consistent with results found elsewhere.[20] No $K\alpha\alpha$ features appear in any of the present solid-target spectra.

A raw spectrum obtained for 1 MeV N^+ ions in solid carbon is shown as curve (a) in Fig. 5. Curve (b) is the same spectrum corrected channel by channel for detector efficiency. This correction is uncertain by +50%, −10% at 1150 eV and by +15%, −5% at 2300 eV. Uncertainties in counting and in solid angle and particle flux measurements are small by comparison. A de-convolution of the (Gaussian) detector response multiplies each point by 0.8 without changing the shape. No

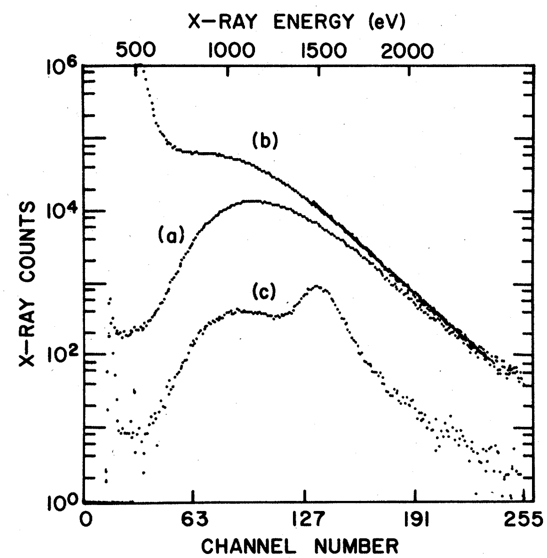

FIG. 5. (a) Si(Li) detector x-ray spectrum (uncorrected) for 1 MeV $N^+ \rightarrow 19\ \mu g/cm^2$ C. (b) The spectrum corrected for detector window absorption. Solid line is a linear least-squares fit to the region beyond the united-atom limit. (c) Si(Li) detector x-ray spectrum (uncorrected) for 1 MeV $N^+ \rightarrow$ C-backed (3.9 $\mu g/cm^2$) Al (19 $\mu g/cm^2$).

correction is made for self-absorption in the thin target.

A semiempirical model for the shape of MO continua in the region beyond the united-atom (UA) limit has been published by Betz *et al.*[21] The shape is predicted to be approximately exponential and we have successfully fitted this spectrum to a single, decaying exponential (solid line in Fig. 5). The spectral half-width is defined as the distance (in eV) between the UA limit E_{UA} and the energy at which the intensity falls to one-half its value at the UA limit. Our fit yields the value 130 eV. The model predicts a half-width of 107 eV that is sensitive to the average value of dE_x/dR where E_x is the MO transition energy and R is the internuclear separation. This derivative can be only crudely estimated since the MO's for this collision system are not known. The model predicts the differential cross section, per incident K-shell vacancy, for spontaneous emission at the UA limit to be

$$\frac{d\sigma^{\text{model}}}{dE_x} = 12 \times 10^{-10}\ \text{Å}^2/\text{eV}$$

for 1 MeV $N^+ \rightarrow C$. The present experimental value,

$$\frac{d\sigma^{\text{expt}}}{dE_x} = 5 \times 10^{-10}\ \text{Å}^2/\text{eV}\ ,$$

believed to be accurate to within a factor of 2, is in reasonable agreement with the model. Possible backgrounds such as bremsstrahlung, radiative electron capture, $K\alpha\alpha$ x rays, and diagram line tails are estimated to be negligible in the collisions studied here. The model considers a two-collision process and takes into account both spontaneous (electric dipole) and (rotationally) induced transitions (not considered here). One-collision processes[22] and two-collision induced transitions, if active, would lead to a discrepancy between the model and the experimental cross sections.

Table I summarizes our measured cross sections and half-widths. It should be emphasized that the present experiment measures the cross section per incident ion, and it does not measure the fraction of projectiles that have K-shell vacancies. That fraction is needed to compare the experimental results with the predictions of the model. To our knowledge, the fractions appropriate to the collision systems studied here have not been published for 1 MeV projectile energy. Alternatively, the fractions can be estimated from ionization and electron-capture cross sections when natural decay can be neglected. Again, the appropriate experimental cross sections have not been published for 1 MeV ions. To obtain the values listed in Table I, col. 3, recently published fractions[23] have been extrapolated from 0.5 to 1 MeV. That extrapolation, especially for O projectiles, has a large uncertainty (perhaps a factor of 2).

TABLE I. Collision-broadening parameters. Entries are for 1 MeV ions and 20 $\mu g/cm^2$ carbon targets. The differential cross section is the value measured at the united-atom limit and is uncertain by a factor of 2. The half-widths are accurate to ±10%.

	$d\sigma/dE$ (10^{-10} Å^2/eV) Expt.[a]			Half-width (eV)	
Ion	(Atom^{-1})	(Vacancy^{-1})	Model[b]	Expt.[a]	Model[b]
C^+	0.4	4	11	131	91
N^+	0.1	5	12	127	96
O^+	0.08	8	14	127	101

[a]This work.
[b]Reference 21 (spontaneous transitions only). These values can vary by 10–20% depending on the particular choices made for model parameters such as atomic K-shell radii and vacancy lifetimes.

A comparison of the $K\alpha\alpha$ yield in the gaseous N_2 target with the MO yield in the solid C target is found by folding the $K\alpha\alpha$ cross section with the detector's Gaussian broadening. The result, at the peak of the spectrum, is

$$\frac{d[K\alpha\alpha]}{dE_x} = 1.3 \times 10^{-12}\ \text{Å}^2/\text{eV}$$

which is $\sim\frac{1}{10}$ the MO cross section measured at the UA limit for 1 MeV N^+ ions in carbon. Furthermore, since K-vacancy sharing favors the lighter collision partner, the N$K\alpha\alpha$ cross section in solid C should be even smaller. The absence of a N$K\alpha\alpha$ peak in our solid target data is therefore attributed to the overwhelming strength of the MO radiation.

IV. SUMMARY

Low-resolution spectra of x rays emitted during collisions of 1–2 MeV C^+, N^+, and O^+ ions with low-Z targets show that gaseous targets yield clean $K\alpha\alpha$ lines, and that these lines can be readily observed for both collision partners.

ACKNOWLEDGMENTS

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