Metastable X-Ray Emitters Produced in Beam-Foil Excitation of Fast Chlorine Beams*

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Metastable x-ray–emitting chlorine ions with lifetimes in the nanosecond region have been found to be produced in the beam-foil excitation of fast chlorine beams. Using coincidence techniques we have determined that these metastable states belong to chlorine ions of two, three, and more than three electrons.

The excitation of the outer shells of fast ion beams by passage through foils is a technique now widely used for producing excitations whose decay is by visible photon emission and whose lifetimes are in the nanosecond region. Inner-shell excitations produced in such a manner are expected to de-excite by x-ray emission and Auger-electron ejection in much shorter times, and, for beams in the MeV/amu region, should generally not live to move more than a few thousand angstroms beyond their excitation point. It has recently been pointed out that this picture is not inclusive, however, and that often metastable states with inner shell vacancies are produced in beam-foil excitation whose lifetimes may be nanoseconds or longer. Sellin *et al.*[1] have reported detection of Auger electrons from metastable quartet states with $1s$ vacancies in three-electron oxygen and fluorine ions following beam-foil excitation of these species. Further, they present evidence for possible metastability of systems with higher electron number. Marrus and Schmieder,[2] using 10-MeV/amu beams, have reported detection of both single- and double-photon emission from foil-excited one- and two-electron metastable states in argon, sulfur, and silicon. Whether the production of metastable K-shell vacancies is confined to three- or fewer-electron systems or is a more general property of foil excitation of all charge states is an interesting question. In this Letter we report the detection of K x rays from metastable two- and three-electron chlorine systems emerging from the foil. We further present experimental evidence that metastable states belonging to systems of higher electron number are produced, indicating that the beam-foil excitation of metastable states with K-shell vacancies is not confined to systems with three or fewer electrons but is a general phenomenon.

We have produced K-shell excitation in highly ionized chlorine ions by passing beams of 20- to 55-MeV ($v/c = 0.035$ to 0.058) chlorine ions through thin (20 $\mu g/cm^2$) carbon foils. At 45 MeV, 80% of the beam emerges from the foil in charge states 11 through 13, and our ability to detect excitations in chlorine systems possessing four to six electrons is favored. We have used a Si(Li) x-ray detector (resolution 165 eV at 3 keV) to view the excited beam at a variable distance downstream from the foil ranging from 0 to 20 cm. An x-ray spectrum taken 2 cm (1.3 nsec) below the foil (Fig. 1) shows $K\alpha$ and $K\beta$ lines from chlorine that are shifted upwards in energy relative to those characteristic of singly ionized chlorine by 145 ± 10 eV and 360 ± 50 eV, respectively, suggesting that their emission is by highly ionized chlorine species. These shifts were found not to depend on the foil-detector separation and were the same as those observed

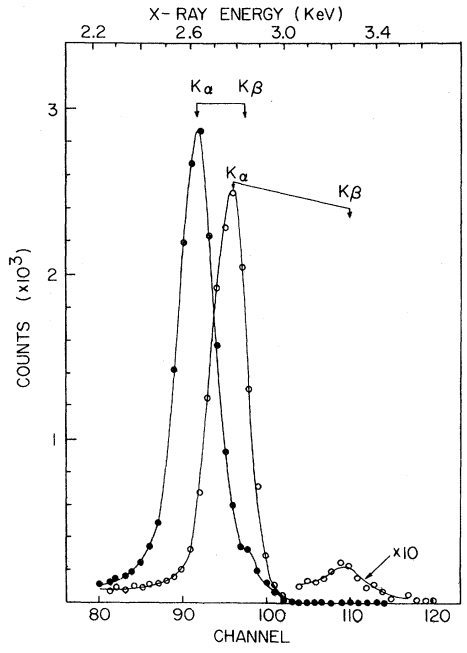

FIG. 1. X-ray energy spectrum from a 45-MeV chlorine beam viewed 2 cm downstream from the exciting foil (open circles). For comparison, an x-ray spectrum produced by proton bombardment of NaCl is shown (closed circles), displaying x-ray energies characteristic of removal of a single $1s$ electron from neutral chlorine.

when the detector was allowed to view the foil directly. Hartree-Fock calculations by House[3] of chlorine $K\alpha$ x-ray energies as a function of the number of remaining M- and L-shell electrons predict shifts of 156 and 134 eV for two- and three-electron chlorine, respectively. One possible conclusion is that the metastable x-ray emitters we observe come from these charge states; however, we will discuss this point further after reviewing the results of our coincidence experiments.

A plot of x-ray yield against foil-detector separation reveals a curve which is not a simple exponential, but which requires at least two exponential components for its description. At a bombarding energy of 35 MeV the data were well described by a sum of decay curves with 1.1- and 9-nsec lifetimes. For comparison, the lifetime associated with a $2p \rightarrow 1s$ transition in hydrogen-like chlorine is 1.9×10^{-14} sec. Although the data could be described by a sum of two exponentials at any bombarding energy, the corresponding lifetime parameters were not energy dependent, indicating that more than two lifetimes are involved. The delayed x-ray yield per incident ion was found to be sharply energy dependent, increasing by 4 orders of magnitude as the bombarding energy was raised from 20 to 55 MeV. Since the fractions of charge states 14–17 emerging from the foil are also increasing rapidly over this energy range, one is again led to suspect that the delayed x rays come from systems of three or fewer electrons.

Our detector resolution of 165 eV is insufficient to allow us any identification of emitting states on the basis of the energy of the radiation alone. We therefore have gone to a coincidence experiment to identify at least the charge state of the metastable emitters. A set of parallel plates operated at 10 kV/cm was used to separate the charge states after x-ray detection. A charge-separated beam of about 10^4 ions/sec was allowed to fall onto the face of a position-sensitive surface-barrier detector in which each ion produced a position signal characteristic of its charge state. The position signal was stored only when it was in delayed coincidence with an x ray in the Si(Li) detector of the appropriate chlorine $K\alpha$ energy. A coincidence resolving time of 70 nsec was used, for which the measured randoms-reals ratio never exceeded 1/1000.

Coincident charge-state spectra were taken separately with the x-ray detector viewing both

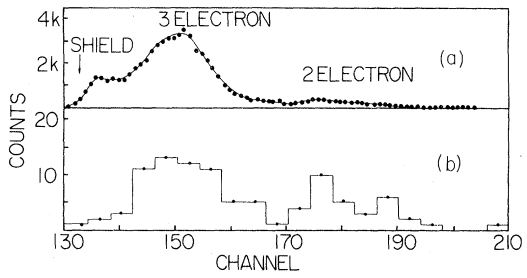

FIG. 2. (a) Singles spectrum from the position-sensitive detector, displaying pulse distributions from charge states 14 and 15, centered near channels 150 and 180, respectively, on the abscissa. Lower charge states were prevented from striking the detector by a shield, whose cutoff appears near channel 134. (b) Corresponding charge-state spectrum of only those ions which were in delayed-coincidence with chlorine K x rays detected 1.5 cm downstream from the foil. The counts are summed over five channels and required 3 h for accumulation.

the foil directly and the excited beam downstream (delayed). In Fig. 2(b) we show the coincident charge-state spectrum taken 1.5 cm (0.95 nsec) after the foil; a corresponding singles spectrum is shown in Fig. 2(a). During the downstream coincidence runs a shield covered the detector in the position of charge states lower than 14 to permit larger counting rates of higher charge states to be obtained. The number of delayed x rays associated with ions belonging to charge states 13 and lower was obtained by subtracting the number of coincidences with higher charge states from the total number of delayed x rays observed as singles, normalized for the collection efficiency of the particle detector. Chlorine $K\alpha$ x rays coming directly from the foil, although having energies characteristic of two- or three-electron systems, were found to be associated with charge states 11 through 15.

In Table I we give approximate values for the total number of delayed x rays per ion for each charge state of the emerging beam, after correcting for appropriate efficiencies. Isotropic x-ray

TABLE I. Approximate total number of delayed K x rays per chlorine ion emerging in a particular charge state from a 45-MeV chlorine beam incident upon a 20-$\mu g/cm^2$ carbon foil.

Charge state	X rays per 1000 ions
15	6 ± 2
14	1.2 ± 0.2
13 and lower	0.3 ± 0.05

emission was assumed, and a universal 1-nsec lifetime was taken to allow us to estimate the fraction of the metastable emitters which decayed within the distance viewed by our x-ray detector. We thus have clear and direct experimental evidence that both two- and three-electron chlorine systems are produced which have lifetimes in the nanosecond region and which de-excite at least partially by $K\alpha$ x-ray emission. We further find that an appreciable fraction (about 60%) of our metastable x-ray emitters belong to systems of more than three electrons. The number of x rays detected from these systems is large because of the charge-state equilibrium at 45 MeV, in spite of the small probability per emerging ion for their production. The appropriate experiment to detect these species directly in a coincidence experiment is in preparation.

The identification of the charge states of these x-ray emitters in no way settles the question of their identity. Sellin *et al.*[1] have attributed their metastable Auger-electron emitters to quartet states in three-electron systems whose autoionization is inhibited by spin selection rules. They further suggest that such systems may also exist for higher electron number, and present possible experimental evidence of their detection. Although a more definite statement must await detailed calculations of the lifetimes expected for such states, we expect that the rapid increase with Z of the spin-orbit mixing of doublet and quartet states will mean that analogous states for $Z=17$ will have lifetimes much shorter than nanoseconds and thus are probably not the states we observe. The consistency of the measured $K\alpha$ x-ray energy with that expected from a two- or three-electron chlorine system, even though more than half of the associated chlorine ions appear to survive with four or more electrons, leads us to suspect that the states seen here may be better attributed to metastable two-electron systems which are accompanied by further electrons residing in shells of very high principal quantum number. Such a system would have to owe its metastability against Auger processes at least in part to the poor overlap of the wave function of the outer-shell electron with that of the inner shells, and to slow radiative progression from outer to inner shells by soft photon emission. Experimental observation of the radiation which should accompany such a progression and deduction of the overall excitation states of the outer shells of foil-excited ions would clearly be of importance in helping such an hypothesis to emerge from the speculation stage.

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Solution of the Dirac Equation for Strong External Fields*

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The 1s bound state of superheavy atoms and molecules reaches a binding energy of $-2mc^2$ at $Z\approx 169$. It is shown that the K shell is still localized in r space even beyond this critical proton number and that it has a width Γ (several keV large) which is a positron escape width for ionized K shells. The suggestion is made that this effect can be observed in the collision of very heavy ions (superheavy molecules) during the collision.

The discrete energy eigenvalues for an electron bound to a nucleus, which are obtained from the Dirac equation, lie between m_0c^2 and $-m_0c^2$, where m_0 is the electron's mass. The problem can be solved analytically in the case of a point nucleus; the energy eigenvalues are then given by the well-known Sommerfeld fine-structure formula. In this case the eigenvalues for the 1s state become imaginary when the nuclear charge Z becomes larger than 137. The problem may be cir-