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Excitation of Swift Heavy Ions in Foil Targets: Visible Radiation from Fe, Ni and Cu

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Abstract

Studies have been made of visible radiation from highly stripped 100–144 MeV Fe, Ni and Cu ions excited by transmission through thin carbon foils. All the observed transitions in the region 180–530 nm were identified with hydrogenic transitions of $\Delta n = 1$ or 2, in charge states between 14 and 22. Light intensities observed are proportional to the outgoing beam charge-state distributions. For thin foils ($< 20\ \mu g\,cm^{-2}$) the light output is less than that from beams in charge equilibrium. It is shown that these observations are consistent with a surface excitation mechanism that depends on the prior formation of multiply excited ions in the solid target.

1. Introduction

This paper describes experiments carried out with the A.N.U. 14 UD accelerator on beam–foil excitation of swift ($\gtrsim 100$ MeV) heavy-ion projectiles ($Z \gtrsim 26$) to study the interactions of these ions in solids. The work described here concerns visible projectile radiation; future studies (to be published elsewhere) will involve X-radiation, and charge exchange and energy losses of such ions.

It is well known that, as a result of transmission through foil targets, heavy ions of 1–10 MeV show *Rydberg* transitions (Lennard *et al.* 1972; Hallin *et al.* 1973; Bashkin 1976; McIntyre *et al.* 1976; Bashkin *et al.* 1978). These transitions are defined (Greenland 1981) as radiative transitions between states of a single electron outside a charged core, and may in general be described by the Ritz series formula with an effective charge q and effective (non-integral!) quantum number n^* (Condon and Shortley 1951). In the limit where the electron orbital radius greatly exceeds that of the core, transitions are *hydrogenic* and, as in hydrogen, may be described by an integral quantum number n with full l degeneracy. In this limit, ion beams of differing atomic number but similar charge states may give rise to identical spectra (Buchet *et al.* 1973). If n is large, the selection rule $\Delta l = \pm 1$ is expected to lead to the depopulation of low-l components by rapid decay to low-n states, so one expects a series of yrast transitions, through the high-l ($=n-1$) states, to occur as a more leisurely sequence. Such sequences are indeed observed, and it is of some interest to understand how high-n states of radii $\approx n^2$ a.u. may be excited by foils. Lennard and Cocke (1973) have argued that a likely mechanism is via electron capture at the foil exit surface. This capture is expected to follow the Oppenheimer (1928) n^{-3} law, as has been observed (Lennard and Cocke 1973; Bukow *et al.* 1976). Confirmation of yrast cascading such as that predicted by Betz (1976) has not been

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directly observed and, of course, its observation would be obscured by the effects of direct feeding and by uncertainties in spectrometer calibrations.

The present work was undertaken to study some of the predictions discussed above, namely the identical spectra of heavy-ion hydrogenic states from different species and the surface foil-excitation mechanism. The A.N.U. Pelletron accelerator is well adapted to produce the beams of different species in high charge states ($q \lesssim 20$) required for such work. The experimental technique, together with the observation and identification of visible beam–foil radiation, are discussed in Section 2. In Section 3 we discuss the known spectra in relation to possible foil-excitation mechanisms. Some data are described on ion charge-state distributions, and their relation to the foil-excitation mechanism is discussed.

2. Experimental Method and Results

(*a*) *Recording of Optical Spectra*

The 14 UD accelerator is able to accelerate Fe, Ni and Cu beams in charge states $q = 9$–11 with currents of $(2\text{–}10) \times 10^{10}$ particles s^{-1} into a target chamber. These beams were passed through self-supporting carbon foils, monitored by a scattered-particle surface barrier detector, and emitted light was observed along a path 2–4 cm downstream of the target. Visible radiation was detected at 90° by a fused silica lens and a dry nitrogen filled McPherson model 218 scanning monochromator, fitted with a 2400 lines per mm grating blazed for 300 mm and a cooled EMI quartz-windowed 6256 S photomultiplier (Carriveau 1970). The area from 2 to 4 cm beyond the target contained a pair of plane electrodes perpendicular to the beam, between which an electric field of 30 kV cm^{-1} could be applied.

The data collection process was similar to that described earlier by Hay and Newton (1980), which involves the counting and recording of output pulses from the photomultiplier, beam charge digitizer, scattered-particle detector and a 100 Hz clock in multiscaling mode with channel advance controlled by the monochromator wavelength readout. The photomultiplier spectrum per 0·5 nm of wavelength was normalized to the number of incident beam particles and allowance was made for the background counting rate of approximately 25 per minute. On account of the low light intensity, the monochromator slits were opened to their maximum extent of 2 mm; the resulting line width of 2·5–3·5 nm was so great that no worthwhile reduction could be obtained by refocusing to cancel the Doppler broadening (cf. Carriveau *et al.* 1972).

Optical spectra were recorded over the wavelength range 180–530 nm at five beam energies between 100 and 144 MeV. Carbon foils with thicknesses between 5 and 60 $\mu g\,cm^{-2}$ were used. In each case the mean charge state of the emergent beam was determined from the Faraday cup readings combined with the known charge of the magnetically selected incident ions. A spectrum taken with 100 MeV Cu ions on a 15 $\mu g\,cm^{-2}$ carbon foil is shown in Fig. 1. This displays a well defined pattern with 'clumps' of several peaks at 30–50 nm spacing and a maximum intensity with minimum width near 350 nm. The data were taken at a measured mean charge state $\bar{q}$ of 19·7 emerging from the foil. Studies at other beam energies showed that the strong central peak altered in shape, and that each clump was thus a blend of several lines with a systematically changing shape within the clump. Moreover, the use of Fe and Ni beams with beam energy having nearly the same $\bar{q}$ led to spectra

almost indistinguishable from that of Fig. 1 (cf. Fig. 2). This observation confirmed the expectation that the optical emission, if hydrogenic, depends directly on the core charge $q+1$ rather than the atomic species. In fact, a detailed description of the lines of Fig. 1 may be made in terms of hydrogenic lines, as discussed in the next subsection.

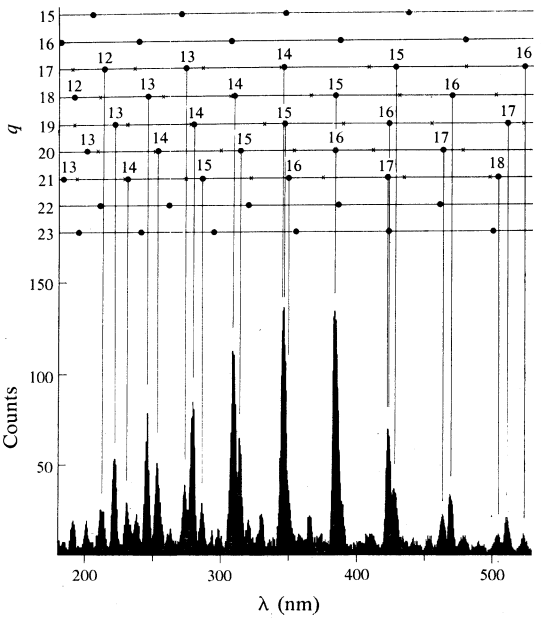

Fig. 1. Spectrum of radiation between 180 and 530 nm from 100 MeV Cu ions after passage through a 15 $\mu g\,cm^{-2}$ carbon foil. Calculated wavelengths for hydrogenic transitions with $\Delta n = 1$ are indicated by solid circles with initial n values. Transitions with $\Delta n = 2$ are indicated by crosses.

(*b*) *Identification of Spectral Lines*

The expected frequency $f = c/\lambda$ for a hydrogenic line with an ion of charge state $q-1$ and principal quantum numbers in an $n \to n'$ transition is*

$$f(q,n,n') = \mathrm{Ry}\, q^2(n'^{-2} - n^{-2}).$$

From this equation one finds that, for lines in the observed light range $\lambda = 180$–530 nm and charge-state range $q = 15$–23, values of n between 12 and 18 contribute. Furthermore, the equation shows that, if a simple relation between n' and n is assumed, there will be near coincidences between certain lines. Thus if $n' = n-1$, the sequence $f(q-2,n-1)$, $f(q,n)$, $f(q+2,n+1)$... is in near coincidence for $n \approx \frac{3}{4}q$ [more precisely one finds $q = 2(n-1)n_+/\{(n+1)n_- - (n+1)n_+\}$, where $n_\pm = (2n \pm 1)^{\frac{1}{2}}$; for $n \gg 1$ the simpler relation follows]. Such a 'piling-up' of lines ('clumps') must enhance the height of the peak at this point. In particular, an enhancement with $q = 19$ at $\lambda = 350$ nm is expected for $n = 15$. This was the starting point of the detailed identification given by the solid circles in Fig. 1, with unique q and n for

* Since the experimental line width was large, fine structure and polarization effects are omitted.

$n' = n-1$, which shows very convincingly the line positions and the way in which the series $f(q-2, n-1)$, $f(q, n)$... 'back-bends' away from the point of coincidence at $\lambda = 350$ nm. Weaker lines for $n' = n-2$ are also identified by crosses. It should be pointed out that there is also a contribution to the fall-off in peak heights above 350 nm by a distinct fall in detector photomultiplier response above 400 nm and a slow decrease of grating efficiency above 300 nm.

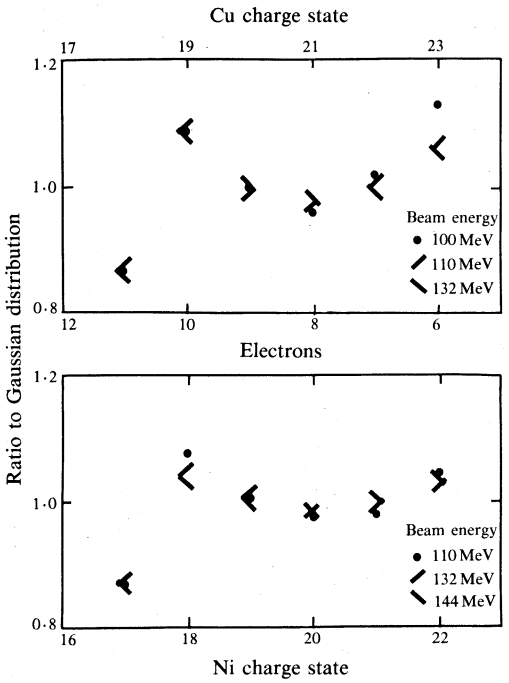

Fig. 2. Relative intensities of lines in Fig. 1 from Cu and from a 20 $\mu g\,cm^{-2}$ Ni target at different beam energies. Data are plotted as deviations from a fitted normal distribution (see text) and are evidently functions of the number of electrons in the different charge states, displaying closure of the L shell at 10 electrons.

Line intensities in Fig. 1 are not in any way governed by the n^{-3} law. This is not surprising since, as discussed in Section 1, the lines observed are predominantly produced from cascading of higher n states. Experimentally, the dominance of cascading in the post-foil region was demonstrated in two ways. First, the application of a 30 $kV\,cm^{-1}$ electric field made no observable difference to the intensity of any line whereas, for mixed-l states, Stark mixing would cause significant quenching with such a field. Secondly, the fall-off in intensity with distance downstream was not more than a factor of 1·5 for 2 cm. This should be compared with that expected for hydrogenic lifetimes for $q = 19$ and $n = 14$–15 (Bromander 1973), namely a factor of 7. From the difference between these values it follows that over 90% of the decays seen 4 cm downstream arise from cascading.

The remark that Fe, Ni and Cu ions at suitable energies give similar spectra can be illustrated by reference to the charge states identified from the spectrum shown in Fig. 1. The intensities of lines from Cu and Ni are plotted in Fig. 2, with q and

n values determined as in Fig. 1. These plots are displayed as deviations from a fitted normal (gaussian) distribution. Small deviations are caused by beam-energy changes, but there is a striking similarity in the data as a function of the residual number of electrons per ion, and in particular in the effect of L-shell closure at 10 electrons ($q = 19$ in Cu and 18 in Ni).

The optical lines of Fig. 1 have thus been shown to depend in a simple way on the mixture of ionic charges present in the beam emerging from the foil. Our description is considerably simpler than the one proposed by Berry and Batson (1976). It is not possible to explain the detailed intensity dependence, however, we shall return to discuss this in Section 3 in relation to the relative charge state distributions of the light and beam.

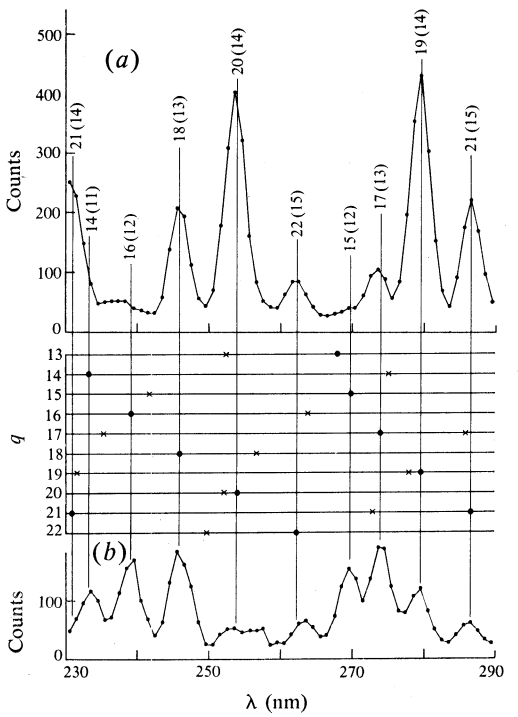

Fig. 3. Beam–foil spectra from 100 MeV Cu ions transmitted through (*a*) a 50 $\mu g\,cm^{-2}$ carbon foil with mean charge state 19·5; (*b*) a 5 $\mu g\,cm^{-2}$ carbon foil with mean charge state 17·0. All the major peaks are for $\Delta n = 1$ transitions, and are labelled numerically by q(initial-n) values. Both curves are normalized to the same number of transmitted beam particles.

(*c*) *Dependence of Light Intensity on Foil Thickness*

The effect of changing foil thickness is illustrated in Fig. 3, which displays data from a range of wavelengths where lines from different charge states are resolved in Fig. 1. Fig. 3*a* shows the spectrum from a 50 $\mu g\,cm^{-2}$ foil, and Fig. 3*b* from a 5 $\mu g\,cm^{-2}$ foil, both curves being normalized to the same number of beam particles. With the thinner target the value of $\bar{q}$ was reduced from 19·5 to 17·0, and this enhanced the transitions involving lower q values. Thus the strongest peaks in

spectrum (*a*) at 253·5 and 279·5 nm, corresponding to $q = 20$ and 19, have almost disappeared in spectrum (*b*), while those at 245·7 and 273·8 nm with $q = 18$ and 17 are now strongest. An important difference in spectrum (*b*) is a significant reduction of the total light output. Fig. 3 displays enough of the visible spectrum to make possible an identification of cascading lines for all the main charge states, so it was possible to measure total light outputs of the two targets as follows. Parts of Fig. 3 containing one transition from the cascade for each charge state were fitted with a Gaussian shape and width corresponding to the spectrometer resolution (with allowance for the background as mentioned in Section 2*a*). For the thick target (Fig. 3*a*), the intensities of $\Delta n = 1$ lines with $q = 17, 19, 21$ ($\approx$280 nm) and 18, 20, 22 ($\approx$250 nm) were found, as a function of q, to have a normal intensity distribution with $\bar{q} = 19{\cdot}5$ and standard deviation $\sigma = 1{\cdot}2$. For the thin target (Fig. 3*b*), lines with $q = 14, 16, 18$ ($\approx$240 nm) and 15, 17, 19, 21 ($\approx$275 nm) were similarly analysed to give $\bar{q} = 16{\cdot}9$ and $\sigma = 2{\cdot}0$, but slightly skewed to the low q side. The counts for the thick and thin targets thus summed were 4527 ± 153 and 2816 ± 151 respectively, i.e. almost a 3 : 2 ratio.

3. Discussion of Excitation Mechanisms

The results of Section 2 show two pieces of information which have some bearing on the processes of foil excitation:

(1) Beam–foil optical radiation appears to come chiefly from yrast transitions with $\Delta n = 1$, cascading down from hydrogenic atoms with physically 'big' radii of order greater than 20^2 a.u.

(2) The light intensity appears to be reduced with smaller excitation foil thickness.

Taken together, these observations would appear to suggest that beam–foil excitation cannot take place directly in the foil or be directly caused at its exit surface; the latter is the usually accepted explanation, based mainly on light-ion data (cf. Garcia 1973; Bickel 1973; Datz 1975). It is therefore important to investigate whether the thickness dependence of the light intensity I reported in Section 2*c* could have been due to the different q, n and λ values for the two targets. If, for instance, a power-law q dependence is assumed then the relation

$$\{\bar{q}(\text{thick})/\bar{q}(\text{thin})\}^a = I(\text{thick})/I(\text{thin})$$

implies, using the values quoted in Section 2*c*, $a = 3{\cdot}3 \pm 0{\cdot}5$. Such a strong q dependence is ruled out by the detailed data of Fig. 1. One might go further and attribute the data to a simultaneous power-law dependence $q^a n^b \lambda^c$. In order to study this numerically, we have compared the relative light and charge fractions in the beam. This comparison led incidentally to a more realistic model of the foil excitation mechanism, as will now be described.

The data of Fig. 1 give the relative light output of the different components of the spectrum, normalized to the number of beam particles. Using an Enge spectrograph, we have measured relative charge fractions, also normalized to the beam. In Table 1 we give the relative light and charge fractions for the different lines defined by q, n and λ in Fig. 1. The virtual constancy of the six values of L/ϕ in Table 1 shows immediately that each power-law exponent a, b or c considered individually must be close to zero. Thus the difference in light outputs of thick and thin foils is not attributable to a q, n or λ dependence.

Table 1. Relative light L and charge ϕ intensities of individual lines shown in Fig. 1

The quantities q, n and λ (nm) are the identified charge state, initial principal quantum number and wavelength respectively

q	n	λ	L/ϕ	q	n	λ	L/ϕ
17	13	274	$1{\cdot}07\pm0{\cdot}07$	20	14	254	$1{\cdot}03\pm0{\cdot}03$
18	13	246	$0{\cdot}97\pm0{\cdot}04$	21	15	286	$0{\cdot}96\pm0{\cdot}04$
19	14	280	$1{\cdot}00\pm0{\cdot}03$	22	15	262	$0{\cdot}97\pm0{\cdot}08$

Our results for the light intensities, given in Table 1 and Fig. 3, show that there is no significant dependence on q, n or λ, and (therefore) that the light intensity shows an increase with foil thickness as the exiting ion charge state distribution approaches equilibrium, as discussed, for example, by Betz (1972). The data of Table 1 also demonstrate accurately that the light intensity is proportional to the charge intensity of the ion producing it. This confirms that the hydrogenic ions are created *subsequent* to passage through the foil: this is not surprising as it is well recognized (cf. Garcia 1973) that such high-n states could hardly survive in solid targets. What is more interesting is that the light intensity per beam increases with the approach to equilibrium of the ion charge distribution, and therefore the creation of hydrogenic atoms is likewise a target-volume effect. It has been shown by Betz and Grodzins (1970) that the approach to charge equilibrium depends on the formation of multiply excited electronic states. It follows that the hydrogenic ions responsible for the observed light are created in these multiply excited ions at (or beyond!) the target exit surface. There is here a striking resemblance to the phenomenon of convoy electron production by ions exiting a solid target: as described by Breinig *et al.* (1982) these convoy electrons are also observed to have intensities proportional to those of the ions with which they emerge in coincidence. Breinig *et al.* have remarked that therefore these electrons are 'produced in the bulk', although more precisely one should say that, like hydrogenic atoms, they are produced in ions which have been *excited* in the bulk.

The electronic processes involved in the production of high-n hydrogenic ions (and similarly of convoy electrons) are not at all understood. There are difficulties, as discussed by Rothermel *et al.* (1982), in attributing their creation to single-electron pick-up at the exit surface. We suggest that it is possible that some form of multiple Auger process such as 'shake-up' (Åberg 1973) may be contributing to the post-foil (vacuum) region. Obviously more measurements on such effects are necessary, and it also seems desirable for studies to be made of the in-target processes such as vacancy production and charge exchange which, apparently, must take place in solid targets prior to the formation of hydrogenic ions and convoy electrons.

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