Charge-State Dependence of the Argon L-Shell Fluorescence Yield Studied by H^+, H_2^+, and He^+ Impact

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The fluorescence yield was determined for the Ar L shell by measuring absolute cross sections for Ar L x-ray and Auger-electron emission for 30- to 600-keV H^+, H_2^+, and He^+ ions on Ar. When the Auger-electron spectrum was analyzed, the fluorescence yield for He^+ was found to be an order of magnitude larger than existing theoretical estimates.

The present work deals with the fluorescence yield of the argon L shell; the vacancies in this shell are created by 30- to 600-keV H^+, H_2^+, and He^+ impact on Ar. The fluorescence yield is defined as the probability that a vacancy in a given shell is filled through a radiative transition from a higher shell. For the Ar L shell the mean fluorescence yield $\bar{\omega}_L$ (further use of the subscript L will be omitted) is much smaller than one, and we can write $\bar{\omega} = \sigma_x/\sigma_A$, where σ_x is the cross section for production of Ar L x rays and σ_A is the cross section for Auger-electron emission. These cross sections have been measured at different laboratories and will be published elsewhere.[1]

Saris and Onderdelinden[2] recognized that for ion-atom collisions the fluorescence yield is dependent both on the projectile ion and the impact energy by comparing their x-ray data for H^+ and Ar^+ on Ar with Auger-electron data.[3,4] More recently, Burch et al.[5] observed an increase of the Ar L fluorescence yield by a factor of ~50 when 30-MeV oxygen ions are used instead of 5-MeV protons. These findings were in qualitative agreement with theoretical estimates by Larkins[6] who used a simple statistical model to predict a significant variation of the Ar L fluorescence yield as a function of outer-shell defects simultaneously produced with inner-shell vacancies. In a more direct calculation, Bhalla and Walters[7] discussed the influence of both ionization and excitation of the outer shell. For more details the reader is referred to current reviews.[8,9]

The present article deals with an extension of the work in Refs. 2–4. In particular, the degree of the simultaneous inner- and outer-shell ionization was evaluated from the intensity of the Auger-satellite lines, and its contribution to the fluorescence yield was calculated using theoretical values[7] for the fluorescence yields of the different configurations. The (semi-) theoretical mean fluorescence yield for the Ar L shell, thus obtained, is compared with the experimental mean fluorescence yield derived from the measured absolute x-ray and Auger-electron production cross sections.

The scattering chamber used for the Auger-electron measurements has been described before,[10] as well as the apparatus for measuring x rays produced by 30- to 135-keV ion impact.[11] The x-ray experiments with 150- to 600-keV ion impact will be published soon.[12]

The results for the experimental mean Ar L fluorescence yield are given in Fig. 1. The measured fluorescence yield is mainly determined by the production of vacancies in the L_{23} shell of argon, including the simultaneous production of defects by ionization or excitation of the M shell. The curves in Fig. 1 show strong variations when different projectile ions are used or when the impact energy is varied.

The measured fluorescence yield is a mean value:

$$\bar{\omega} = \sum_i q_i \omega(L_{23}M^i) \quad (i = 0, 1, 2, \ldots),$$

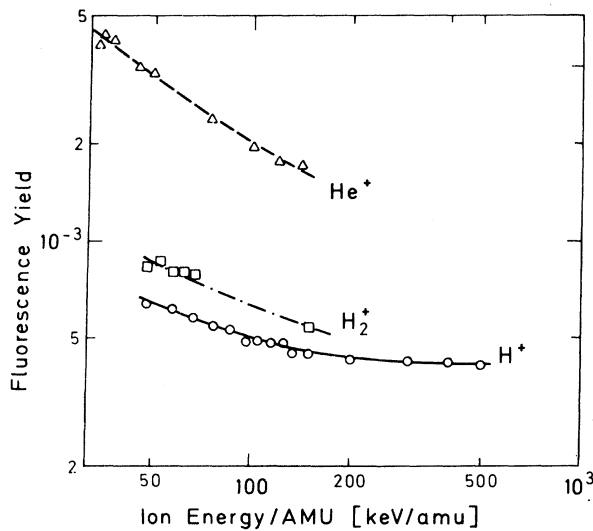with $\sum_i q_i = 1$, where q_i is the relative probability

FIG. 1. Mean fluorescence yield for the Ar L shell obtained by H^+, H_2^+, and He^+ impact excitation. The error of the absolute values is about 20%.

FIG. 2. Ar L_{23} Auger spectra produced by 75-keV H^+ and 300-keV He^+ impact. Energies of the level diagram are taken from Refs. 14 and 15.

for the production of an $L_{23} M^i$ configuration and $\omega(L_{23}M^i)$ is the corresponding fluorescence yield; $L_{23}M^i$ stands for a configuration with an L_{23} vacancy and a number i of M-shell vacancies produced by ionization and excitation. The q_i imply also the formation of L_1 defects, as they are mainly converted to L_{23} vacancies by Coster-Kronig transitions.[13]

Because in our example the fluorescence yield is always much smaller than 1, the Auger-electron yield is essentially equal to 1, and the q_i values can be derived from the Ar L_{23} Auger spectrum (see Fig. 2). Although the individual satellite lines were often not resolved, the relevant q_i values could be evaluated from the four main peaks of the Auger spectrum. Normal Auger transitions contribute to three of these peaks[14] according to the final states $M_1 M_1$, $M_1 M_{23}$, and $M_{23} M_{23}$. The Auger peaks are simultaneously shifted to lower energies as an additional outer-shell vacancy occurs.[15] The value of q_0 was determined in the following way: The pure diagram peak at 205 eV was integrated, and branching ratios[14] for normal transitions were used to calculate the fraction of the normal transitions in the other peaks; dividing the sum of all normal lines by the integral of the whole spectrum gives q_0. After a subtraction of the normal-line fractions from the Auger spectrum, q_1 was evaluated by an analogous procedure as for q_0. The branch-
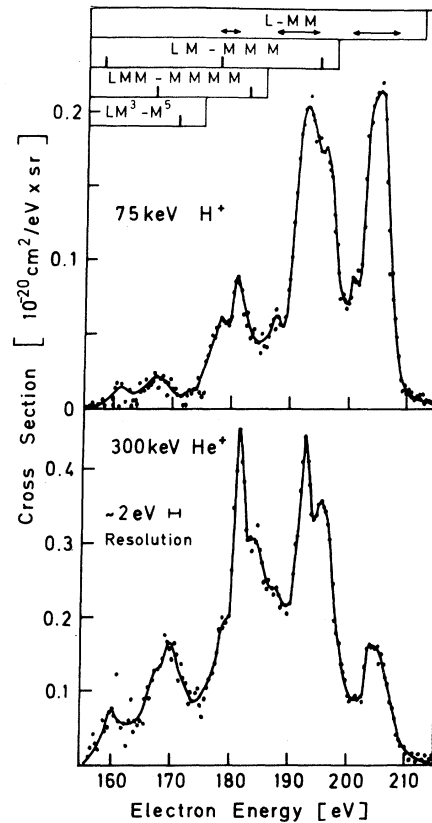
ing ratios for this case were determined from the satellite spectra obtained with 400- to 600-keV protons on argon. In this energy range the spectra appear to contain only a few-percent higher-order $(i > 1)$ satellites. After a subtraction of the q_1-satellite values the remainder was attributed to q_2. The subtraction method applies well for He^+ impact where q_2 is relatively large. For H^+ and H_2^+ impact q_2 was estimated from the intensity of the 181-eV satellite which contributes mainly to q_2. In Table I are shown some results of the q-value analysis.

Using the experimental q_i and theoretical $\omega(L_{23}-M^i)$ values, the (semi-) theoretical mean fluorescence yield $\bar{\omega}_{\text{theor}} = \sum q_i \omega(L_{23}M^i)$ was derived and compared to the experimental mean fluorescence yield $\bar{\omega}_{\text{expt}}$ (see Table I). We have chosen $\omega(L_{23}-M^i) = (2.0, 2.6, 3.7) \times 10^{-4}$ for $i = 0, 1, 2$, respectively, as calculated by Bhalla and Walters[7] for $L_{23}-M_{23}{}^i$ defect configurations, where a number i of

TABLE I. Relative cross sections q_i for the production of $L_{23}M^i$ config-
urations in Ar by H^+, H_2^+, and He^+ impact. Errors for q_0, q_1, and q_2
(for He^+) are about $\pm 10\%$, $\pm 20\%$, and $\pm 30\%$, respectively. $\bar{\omega}_{theor}$, $\bar{\omega}_{fit}$,
and $\bar{\omega}_{expt}$ are (semi-) theoretical, empirical, and experimental fluores-
cence yields.

Ion	Energy [keV]	q_0	q_1	q_2	$\bar{\omega}_{theor}$ x 10^4	$\bar{\omega}_{fit}$ x 10^4	$\bar{\omega}_{exp}$ x 10^4
H^+	500	0.59	0.39	∿0.02	2.25	4	4.1 ± 0.8
	150	0.47	0.51	∿0.025	2.35	4.5	4.5 ± 0.9
	75	0.36	0.61	∿0.03	2.45	5	5.5 ± 1
H_2^+	300	0.41	0.55	∿0.035	2.4	6	5.9 ± 1
	150	0.31	0.64	∿0.05	2.5	7	7.2 ± 1.5
	100	0.29	0.64	∿0.06	2.55	8	8.6 ± 1.5
He^+	600	0.20	0.63	0.16	2.6	17	16 ± 3
	300	0.14	0.59	0.26	2.75	26	24 ± 5
	135	0.075	0.56	0.36	2.95	36	43 ± 8

M_{23} electrons are removed to continuum. In ad-
dition to these configurations which are expected
to contribute mainly to a certain q_i, there are
initial configurations with defects in the M_1 shell.
Furthermore, configurations occur where elec-
trons from both the M_{23} and the M_1 shells are
excited to discrete states. Bhalla and Walters[7]
have calculated that the fluorescence yield in-
creases when a $3p$ electron is excited to a $3d$
state instead of being removed to the continuum.
Larkins[6] has shown that defects in the M_1 shell
generally decrease the fluorescence yield. Thus,
the numbers for $\omega(L_{23}M^i)$ chosen above appear
as reasonable choices of the theoretical fluores-
cence yields related to the q_i.

The results in Table I show considerable dis-
crepancies between the theoretical and experi-
mental fluorescence yields, in particular for He^+
impact. To fit the experimental data, a mean
fluorescence yield $\bar{\omega}_{fit}$ (shown in Table I) was de-
duced from empirical $\omega(L_{23}M^i)$ values. A good
overall agreement was obtained by leaving the
theoretical $\omega(L_{23}M^0)$ and $\omega(L_{23}M^1)$ unchanged and
replacing $\omega(L_{23}M^2)$ by the comparatively large
value of 95×10^{-4}. The fit procedure shows that
the number of $\omega(L_{23}M^2)$ is relatively well estab-
lished; it follows directly from the analogous
variation of $\bar{\omega}_{expt}$ and q_2 with the energy of He^+.
As the $q_2\omega(L_{23}M^2)$ term is about 90% for He^+,

the value of $\omega(L_{23}M^2)$ is nearly independent of
the choices of $\omega(L_{23}M^0)$ and $\omega(L_{23}M^1)$. The lat-
ter values cannot be uniquely determined in the
fit procedure; however, the experimental data
for H^+ and H_2^+ show at least that they have ap-
proximately the magnitude theoretically predict-
ed. When choosing the theoretical values given
by Bhalla and Walters[7] for $\omega(L_{23}M^0)$ and $\omega(L_{23}-M^1)$, the experimental results for H^+ and H_2^+
are fitted well by the calcultated values (see
$\bar{\omega}_{fit}$ in Table I).

The precise origin of the large $\omega(L_{23}M^2)$ value
is not known. The analogous variation of $\bar{\omega}_{expt}$
and the q_2 value suggests that the large fluores-
cence yield observed in particular for He^+ impact
is mainly caused by outer-shell configurations
resulting in q_2 satellites. The q_2 value is mainly
determined by one satellite line at 181 ± 0.4 eV
which should be attributed to a $L_{23}M^2$ defect con-
figuration (see Fig. 2). On the other hand, it is
not expected that the fluorescence yield for an
$L_{23}M^2$ defect configuration is an order of magni-
tude larger than the theoretical value. Probably
other effects in addition to the charge-state in-
fluence must be taken into account.

Mehlhorn[16] suggested that the number of radia-
tive transitions to the L_1 shell is comparatively
large for highly ionized atoms when the Coster-
Kronig transitions are absent. Preliminary hfs

calculations[16] show that Coster-Kronig transitions are energetically forbidden when more than two vacancies occur in the outer shell. In the absence of competing Coster-Kronig transitions the fluorescence yield of the L_1 shell increases by more than an order of magnitude. For the present case we estimated that the number of highly ionized atoms $(i > 2)$ is too small for a significant effect from the L_1 shell, but more detailed work is needed to study the charge-state dependence of the fluorescence yield and the Coster-Kronig transition rate.

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Energetic Heavy-Particle Detection by Photochromic Material

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Mechanisms are discussed by which radiations can produce color changes in Fe-doped $SrTiO_3$. α-particle and cosmic-ray (π-meson) absorption events have been detected by these crystals.

We report here the detection of high-energy particles by photochromic iron-doped strontium-titanate single crystals. This method is much simpler than detection by other presently existing track-type detectors. As explained below, the tracks can be erased and the crystal may be reused.

Strontium-titanate crystals doped with 0.1% iron are black in color. The iron ions enter the crystal substitutionally, replacing Ti^{4+}. About half of the iron ions ($\sim 52\%$) are in the Fe^{4+} state.[1]

The black color is explained in terms of the high absorption coefficient of Fe^{4+} in the visible region of the spectrum.[2] If such a crystal is reduced by heating at 900°C for about $\frac{1}{2}$ hour in partial vacuum (1 Torr), the number of Fe^{3+} ions increases and the crystal changes color from black to yellow. Since the Fe^{3+} ions are in the Ti^{4+} sites, charge balance in the crystal is maintained by oxygen vacancies.[3] By changing the valence state of the iron ions, the color of the crystal can be switched back and forth between black and