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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 Onderdelinden² 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 Larkins⁶ 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 Walters⁷ 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 values⁷ 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:

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

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

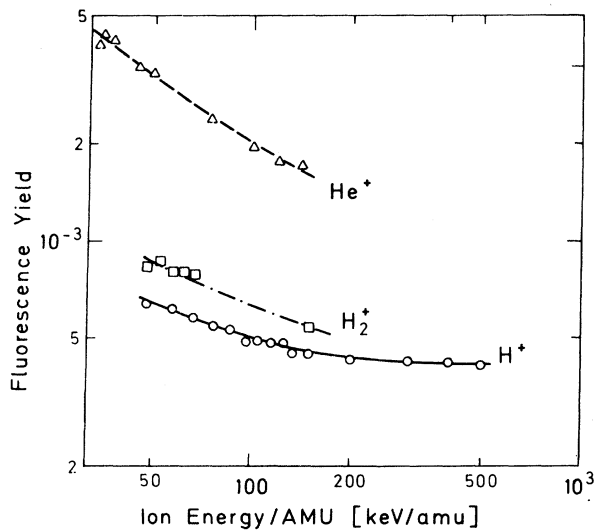


FIG. 1. Mean fluorescence yield for the Ar L shell obtained by H^+ , H_2^+ , and He^+ impact excitation. The error of the absolute values is about 20%.

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 peaks¹⁴ 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 ratios¹⁴ 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-

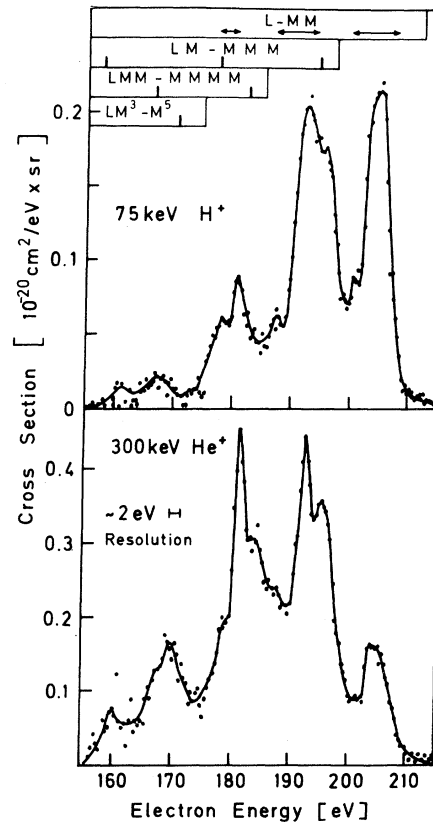


FIG. 2. Ar L_{23} Auger spectra produced by 75-keV H^+ and 300-keV He^+ impact. Energies of the level diagram are taken from Refs. 14 and 15.

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 Walters⁷ for $L_{23} M_{23}^i$ defect configurations, where a number i of

TABLE I. Relative cross sections q_i for the production of $L_{23}M^i$ configurations 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. $\bar{\omega}_{\text{theor}}$, $\bar{\omega}_{\text{fit}}$, and $\bar{\omega}_{\text{expt}}$ are (semi-) theoretical, empirical, and experimental fluorescence yields.

Ion	Energy [keV]	q_0	q_1	q_2	$\bar{\omega}_{\text{theor}}$ $\times 10^4$	$\bar{\omega}_{\text{fit}}$ $\times 10^4$	$\bar{\omega}_{\text{exp}}$ $\times 10^4$
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 addition 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 electrons from both the M_{23} and the M_1 shells are excited to discrete states. Bhalla and Walters⁷ have calculated that the fluorescence yield increases when a $3p$ electron is excited to a $3d$ state instead of being removed to the continuum. Larkins⁶ 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 fluorescence yields related to the q_i .

The results in Table I show considerable discrepancies between the theoretical and experimental fluorescence yields, in particular for He^+ impact. To fit the experimental data, a mean fluorescence yield $\bar{\omega}_{\text{fit}}$ (shown in Table I) was deduced 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 established; it follows directly from the analogous variation of $\bar{\omega}_{\text{expt}}$ 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 latter values cannot be uniquely determined in the fit procedure; however, the experimental data for H^+ and H_2^+ show at least that they have approximately the magnitude theoretically predicted. When choosing the theoretical values given by Bhalla and Walters⁷ 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 calculated values (see $\bar{\omega}_{\text{fit}}$ in Table I).

The precise origin of the large $\omega(L_{23}M^2)$ value is not known. The analogous variation of $\bar{\omega}_{\text{expt}}$ and the q_2 value suggests that the large fluorescence 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 configuration (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 magnitude larger than the theoretical value. Probably other effects in addition to the charge-state influence must be taken into account.

Mehlhorn¹⁶ suggested that the number of radiative transitions to the L_1 shell is comparatively large for highly ionized atoms when the Coster-Kronig transitions are absent. Preliminary hfs

calculations¹⁶ 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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¹X-ray data: K. G. Harrison, H. Tawara, and F. J. de Heer, to be published; A. Langenberg and J. van Eck, to be published [using the same technique as F. W. Saris and D. Onderdelinden, *Physica (Utrecht)* **49**, 441 (1970)]. Auger data: N. Stolterfoht, D. Schneider, and P. Ziem, to be published.

²Saris and Onderdelinden, Ref. 1.

³D. J. Volz and M. E. Rudd, *Phys. Rev. A* **2**, 1395 (1970).

⁴R. K. Cacak, G. C. Kessel, and M. E. Rudd, *Phys. Rev. A* **2**, 1327 (1970).

⁵D. Burch, W. B. Ingalls, J. S. Risley, and R. Heffner, private communication.

⁶F. P. Larkins, *J. Phys. B: Proc. Phys. Soc., London* **4**, L29 (1971).

⁷C. P. Bhalla and K. L. Walters, in *Proceedings of the International Conference on Inner Shell Ionization Phenomena, Atlanta, Georgia, 1972*, edited by R. W. Fink, S. T. Manson, I. M. Palms, and P. V. Rao, CONF-720 404 (U. S. Atomic Energy Commission, Oak Ridge, Tenn., 1973), p. 1572.

⁸*Proceedings of the International Conference on Inner Shell Ionization Phenomena, Atlanta, Georgia, 1972*, edited by R. W. Fink, J. T. Manson, I. M. Palms, and P. V. Rao, CONF-720 404 (U. S. Atomic Energy Commission, Oak Ridge, Tenn., 1973).

⁹F. W. Saris, in *Proceedings of the Seventh International Conference on the Physics of Electronic and Atomic Collisions. Invited Talks and Progress Reports*, edited by T. R. Govers and F. J. de Heer (North-Holland, Amsterdam, 1972).

¹⁰N. Stolterfoht, *Z. Phys.* **248**, 81, 92 (1971).

¹¹K. G. Harrison, H. Tawara, and F. J. de Heer, to be published.

¹²A. Langenberg and J. van Eck, to be published.

¹³W. Mehlhorn, *Z. Phys.* **208**, 11 (1968).

¹⁴W. Mehlhorn and D. Stahlherm, *Z. Phys.* **217**, 294 (1968).

¹⁵F. P. Larkins, *J. Phys. B: Proc. Phys. Soc., London* **4**, 1 (1971).

¹⁶W. Mehlhorn, private communication.

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 re-used.

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