Charge-State Dependence of the Argon L-Shell Fluorescence Yield Studied by H+, H2+, and He+ Impact

Stolterfoht, N.; de Heer, F.J.; Eck, J.van

Published in:
Physical Review Letters

DOI:
10.1103/PhysRevLett.30.1159

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

N. Stolterfoht
Abteilung Strahlenphysik, Hahn-Meitner-Insitut für Kernforschung Berlin GmbH, Berlin-West, Germany

and

F. J. de Heer
FOM-Instiuut voor Atoom-en Molecuulfysica, Amsterdam/Watergraafsmeer, The Netherlands

and

J. van Eck
Rijksuniversiteit Utrecht, Fysisch Laboratorium, Utrecht, The Netherlands
(Received 20 March 1973)

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

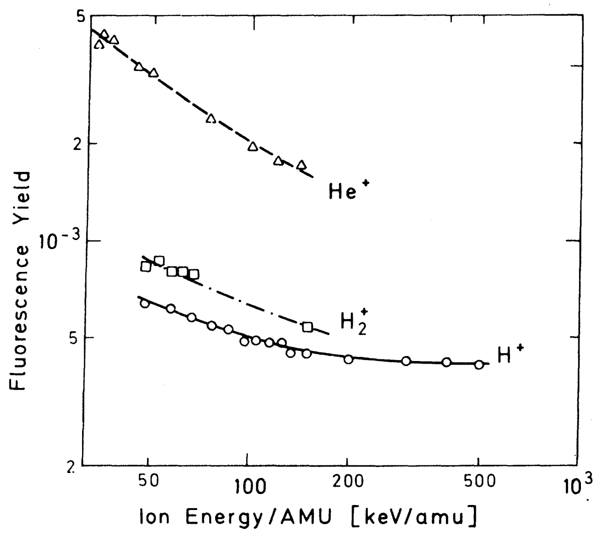

FIG. 1. Mean fluorescence yield for the Ar L shell obtained by H^+, H_2^+, and He^+ impact excitation. The error of the absolute values is about 20%.

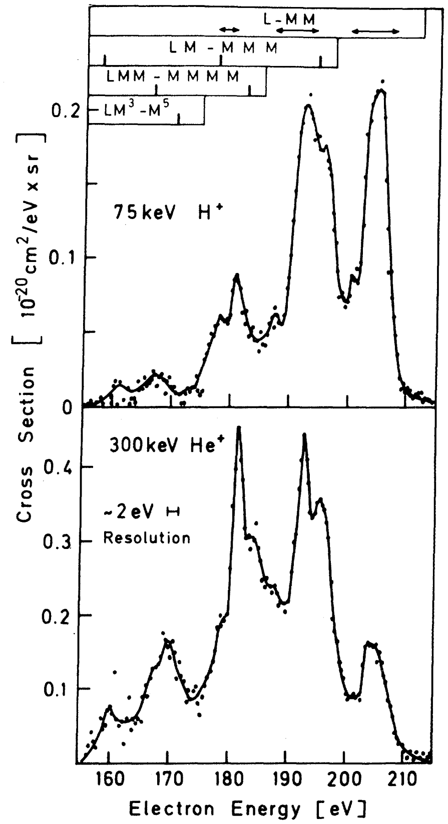

FIG. 2. Ar L_{23} Auger spectra produced by 75-keV H^+ and 300-keV He^+ impact. Energies of the level diagram are taken from Refs. 14 and 15.

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_1M_1, M_1M_{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 branching 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 $L_{23}M^i$ configurations in Ar by H^+, H_2^+, and He^+ impact. Errors for q_0, q_1, and q_2 (for He^+) are about ±10%, ±20%, and ±30%, respectively. $\bar{\omega}_{theor}$, $\bar{\omega}_{fit}$, and $\bar{\omega}_{expt}$ are (semi-) theoretical, empirical, and experimental fluorescence yields.

Ion	Energy [keV]	q_0	q_1	q_2	$\bar{\omega}_{theor} \times 10^4$	$\bar{\omega}_{fit} \times 10^4$	$\bar{\omega}_{exp} \times 10^4$
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 Walters[7] have calculated that the fluorescence yield increases 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 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 $\bar{\omega}_{fit}$ (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 $\bar{\omega}_{expt}$ 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 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 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.

Mehlhorn[16] 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 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.

We are very much indebted to Dr. H. Tawara, Dr. K. G. Harrison, Mr. A. Langenberg, Mr. D. Schneider, and Mr. U. Leithäuser who shared in the experimental work. We would like to thank Dr. D. Burch and colleagues for providing us with their experimental results prior to publication, and Professor Dr. W. Mehlhorn for very helpful sugestions. Part of this work is sponsored by FOM with financial support from ZWO.

[1]X-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.

[2]Saris and Onderdelinden, Ref. 1.

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

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

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

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

[7]C. 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).

[9]F. 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).

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

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

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

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

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

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

[16]W. Mehlhorn, private communication.

Energetic Heavy-Particle Detection by Photochromic Material

P. J. Ouseph
Physics Department, University of Louisville, Louisville, Kentucky 40208
(Received 20 February 1973)

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 (~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