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Determination of metastable fractions in noble-gas-ion beams

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Yields and kinetic energy distributions of secondary electrons emitted by potential emission due to impact of slow singly or multiply charged noble-gas ions on a clean polycrystalline tungsten target have been determined. An empirical relation for the secondary-electron yield of ground-state ions is applied to obtain the secondary-electron yield of corresponding metastably excited ions. By this technique metastable fractions of noble-gas-ion beams could be evaluated. Results for singly and doubly charged ions of Ar, Kr, and Xe, are presented.

I. INTRODUCTION

Nowadays, beams of singly and multiply charged ions are widely used in experimental atomic physics and for investigation of fundamental plasma processes. In principle, such studies involve the interaction of ions with atomic particles or surfaces. Whenever possible, noble-gas ions are preferred because they can be produced and handled most conveniently. To assure clean experimental conditions the ions should be well defined according to their mass, charge state, and kinetic energy. However, possibly present admixtures of metastably excited ions surpass these procedures and may cause considerable experimental inconveniencies. On the other hand, investigations which are directly related to metastable ions may be of interest, too.

Although various methods to demonstrate the presence of metastable particles in ion beams are known, only in a few special cases could the quantitative determination of metastable ion beam fraction be achieved so far. Hagstrum[1,2] has demonstrated that metastable singly charged noble-gas ions manifest themselves in the emission of electrons from solid surfaces as induced by these slow ions [potential emission (PE)]. Metastable ion beam fractions were estimated by assuming the electron yield of metastable ions, $\gamma_{1,m}$ being equal to the electron yield of doubly charged ground-state ions γ_2. For the fraction f of metastable ions this yields[1]

$$f \simeq \gamma_{\text{eff}} - \gamma_1/\gamma_2 - \gamma_1 . \qquad (1)$$

γ_{eff} denotes the electron yield as measured for ion beams with unknown metastable admixtures. γ_1 and γ_2, the electron yields for singly and doubly charged ground-state ions, respectively, have been measured with appropriately prepared ion beams.

The experimental results for impact of singly charged ground-state noble-gas ions on clean metal surfaces have been discussed within the theory of PE developed by Hagstrum.[3] Both yield and kinetic energy distribution $K(E)$ could be explained in detail. We have carried out similar PE investigations as an attempt to determine quantitatively metastable ion fractions in beams of both singly and multiply charged noble-gas ions.

II. EXPERIMENTAL

Singly or multiply charged noble-gas ions (ion charge number $z = 1, 2, 3$) were extracted from an ion source with controllable electron bombardment energy E_{IQ}. The ions were analyzed according to mass and charge state, decelerated to energies E_i adjustable from 20 eV up to several hundred eV, and were directed onto a polycrystalline W target, which was cleaned regularly by both flash desorption and glow-discharge sputtering. Typically, ion currents at the target were of the order of 10^{-10} A. The investigations were carried out with an essentially clean target at a background pressure around 10^{-8} Pa. The electron yield γ was determined from measurements of both target current and the current of secondary electrons emitted from the target. The latter was measured with a hemispherical collector surrounding the target. To investigate the electron kinetic energy distribution $K(E)$ up to three hemispherical retarding grids were introduced into the collector assembly and transmitted electrons were detected by means of a channel electron multiplier mounted behind a small aperture in the collector. A detailed description of the apparatus and the experimental procedures is given elsewhere.[4] Experimental uncertainties of the obtained γ-values are below ± 5%.

III. RESULTS

Within the quoted error limits, our γ data for singly and multiply charged ground-state ions agree with those given by Hagstrum.[5,6] Table I

TABLE I. γ_z values of 100-eV ground-state noble-gas ions impinging on clean polycrystalline W.

z	Ion	γ_z(from Ref. 4)	γ_z(from Refs. 5 and 6)
1	Xe^+	0.013	0.012
	Kr^+	0.049	0.051
	Ar^+	0.090	0.095
	Ne^+	0.245	0.246
	He^+	0.26	0.263
2	Xe^{+2}	0.21	0.22
	Kr^{+2}	0.31	0.30
	Ar^{+2}	0.36	0.38
	Ne^{+2}	0.68	0.70
	He^{+2}	⋯	0.72
3	Xe^{+3}	⋯	0.77
	Kr^{+3}	0.90	0.90
	Ar^{+3}	1.15	1.20
	Ne^{+3}	⋯	1.80
4	Xe^{+4}	⋯	1.65
	Kr^{+4}	⋯	1.95
5	Xe^{+5}	⋯	2.90

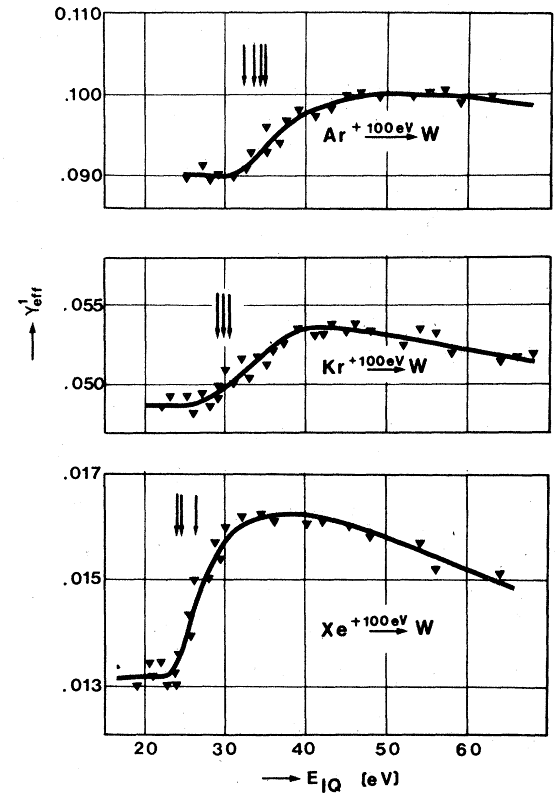

FIG. 1. γ_{eff} data for singly charged noble-gas-ion beams with metastable-ion admixtures dependent on E_{IQ}. Appearance potentials for metastable states indicated by arrows. Ion impact energy $E_i = 100$ eV.

shows our results for ion impact energies of $E_i = 100$ eV. In Figs. 1 and 2 measurements of γ_{eff} versus E_{IQ}, the electron bombardment energy in the ion source, are shown.

For singly charged ions our results resemble those of Hagstrum[1] with the difference that at higher E_{IQ} still no influence due to doubly charged ions appeared because of our different experimental arrangement. For the first time, doubly charged metastable ions are demonstrated by means of the PE method.

In Table II the known metastable states of singly and doubly charged noble-gas ions and their respective excitation energies above the ion ground states $W_{z,m}$ are listed. Data were taken from Refs. 7 and 8. The appearance potentials for the various metastable states have been indicated by arrows in Figs. 1 and 2.

It is known that by electron bombardment only minute metastable ion beam fractions are achievable for He^+ ions[1] as well as for Ne^+ ions.[9] Therefore, we have restricted our investigations mainly to the heavier noble-gas ions.

We have also determined the kinetic energy distributions $K(E)$ of secondary electrons. Of special interest is the high-energy part of the respective $K(E)$. The retardation technique yields integrated values of $K(E)$, cf. Fig. 3.

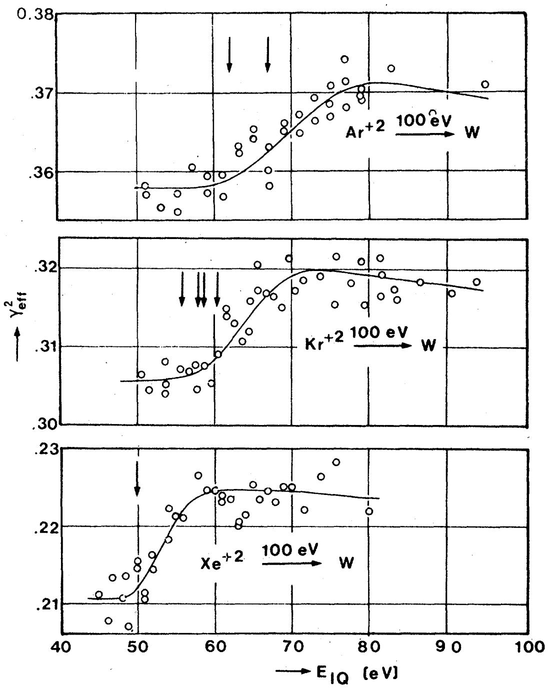

FIG. 2. γ_{eff} data for doubly charged noble-gas-ion beams with metastable-ion admixtures dependent on E_{IQ}. Appearance potentials for metastable states indicated by arrows. Ion impact energy $E_i = 100$ eV.

TABLE II. Metastable states of singly and doubly charged noble-gas ions. $W_{z-1,i}$ is the ion recombination energy; $W_{z,m}$ is the metastable ion excitation energy; $A_z, A_{z,m}$ are the appearance potentials.

		Ground state			Metastable states	
Ion	Term	$W_{z-1,i}$ (eV)	A_z (eV)	Term	$W_{z,m}$ (eV)	$A_{z,m}$ (eV)
Xe^+	$5p^5\ {}^2P^o_{3/2}$	12.1	12.1	$5p^5\ {}^2P^o_{1/2}$	1.3	13.4
				$5d\ {}^4D_{7/2}$	11.8	24.0
				$5d\ {}^4F_{7/2}$	12.3	24.4
				$5d\ {}^4F_{9/2}$	12.3	24.5
				$5d\ {}^2F_{7/2}$	14.3	26.4
Kr^+	$4p^5\ {}^2P^o_{3/2}$	14.0	14.0	$4p^5\ {}^2P^o_{1/2}$	0.7	14.7
				$4d\ {}^4D_{7/2}$	14.9	28.9
				$4d\ {}^4F_{9/2}$	15.6	29.6
				$4d\ {}^4F_{7/2}$	15.9	29.9
				$4d\ {}^2F_{7/2}$	16.3	30.3
Ar^+	$3p^5\ {}^2P^o_{3/2}$	15.8	15.8	$3p^5\ {}^2P^o_{1/2}$	0.2	15.9
				$3d\ {}^4D_{7/2}$	16.4	32.2
				$3d\ {}^4F_{9/2}$	17.6	33.4
				$3d\ {}^4F_{7/2}$	17.7	33.5
				$3d\ {}^2F_{7/2}$	18.5	34.3
				$3d'\ {}^2G_{9/2}$	19.1	34.9
				$3d'\ {}^2G_{7/2}$	19.1	34.9
Ne^+	$2p^5\ {}^2P^o_{3/2}$	21.6	21.6	$2p^5\ {}^2P^o_{1/2}$	0.1	21.7
				$3s\ {}^4P_{5/2}$	27.2	48.8
				$3s\ {}^4P_{3/2}$	27.2	48.8
				$3s\ {}^4P_{1/2}$	27.3	48.9
He^+	$1s\ {}^2S_{1/2}$	24.6	24.6	$2s\ {}^2S_{1/2}$	40.8	65.4
Xe^{+2}	$5p^4\ {}^3P_2$	21.2	33.3	$5p^4\ {}^3P_0$	1.0	34.4
				$5p^4\ {}^3P_1$	1.2	34.6
				$5p^4\ {}^1D_2$	2.1	35.5
				$5p^4\ {}^1S_0$	4.6	38.0
				$5d'\ {}^3G^o_4$	16.4	49.7
				$5d'\ {}^1G^o_4$	16.4	49.8
Kr^{+2}	$4p^4\ {}^3P_2$	24.6	38.6	$4p^4\ {}^3P_1$	0.6	39.1
				$4p^4\ {}^3P_0$	0.7	39.2
				$4p^4\ {}^1D_2$	1.8	40.4
				$4p^4\ {}^1S_0$	4.1	42.7
				$4d\ {}^5D^o_4$	17.2	55.8
				$4d'\ {}^3F^o_4$	19.4	57.9
				$4d'\ {}^3G^o_4$	19.9	58.5
				$4d'\ {}^3G^o_5$	20.0	58.5

TABLE II. *(Continued)*.

Ion	Term	Ground state $W_{z-1,i}$ (eV)	A_z (eV)	Metastable states Term	$W_{z,m}$ (eV)	$A_{z,m}$ (eV)
				$4d'\ ^1G_4^o$	20.2	58.8
				$4d''\ ^3F_4^o$	21.7	60.3
Ar^{+2}	$3p^4\ ^3P_2$	27.6	43.4	$3p^4\ ^3P_1$	0.1	43.5
				$3p^4\ ^3P_0$	0.2	43.6
				$3p^4\ ^1D_2$	1.7	45.1
				$3p^4\ ^1S_0$	4.1	47.5
				$3d\ ^5D_4^o$	18.0	61.4
				$3d'\ ^3F_4^o$	23.1	66.5
Ne^{+2}	$2p^4\ ^3P_2$	41.1	62.6	$2p^4\ ^3P_1$	0.1	62.7
				$2p^4\ ^3P_0$	0.1	62.8
				$2p^4\ ^1D_2$	3.2	65.8
				$2p^4\ ^1S_0$	6.9	69.6
				$3s\ ^5S_2^o$	39.0	101.6

IV. DISCUSSION

For ground-state ions X^{+z} the theory of PE[3] predicts for $K(E)$ the maximum energy

$$E^{z}_{\max} = W'_{z-1,i} - 2W_{\phi}, \qquad (2)$$

where $W_{z-1,i}$ is the ionization potential of $X^{+(z-1)}$ and W_{ϕ} is the work function of the target surface. It is assumed that X^{+z} becomes neutralized in a sequence of processes that can be both of the Auger neutralization and the resonance-neutralization–Auger-deexcitation type. The influence of level shifts in the approaching particle is indicated by a prime. In most cases the levels will shift upwards which results in a slight decrease of $E^{z}_{\max}$.[3,10]

For metastably excited ions, as the first process, most probably Auger deexcitation will take place. There, the maximum electron energy can amount to

$$E^{z,m}_{\max} = W'_{z,m} - W_{\phi}. \qquad (3)$$

Again, possible effects due to level shifts have been indicated by a prime. Comparison of (2) with (3) shows that the presence of metastable ion-beam admixtures can manifest itself in the high-energy part of $K(E)$ only as long as

$$E^{z,m}_{\max} > E^{z}_{\max}. \qquad (4)$$

However, the electron yield will be modified by metastable admixtures as soon as

$$W'_{z,m} > W_{\phi}. \qquad (5)$$

For beams of Ar^{+} and Kr^{+}, respectively, the in-

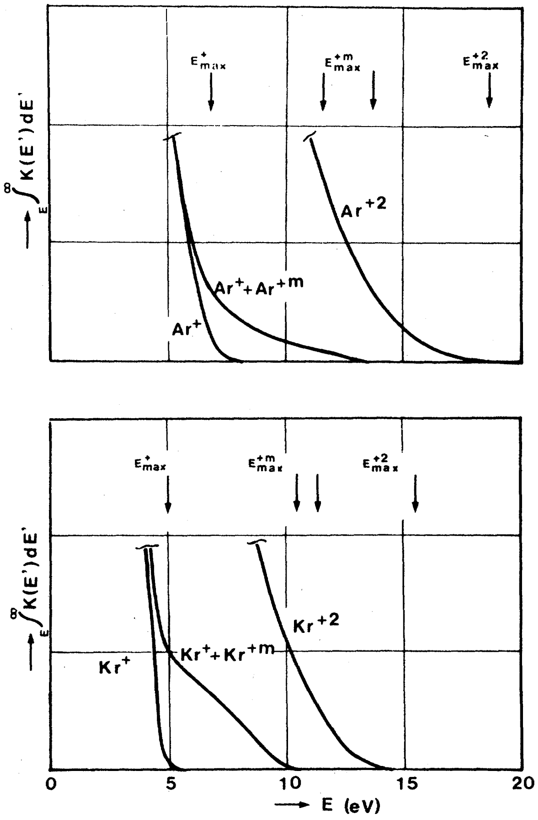

FIG. 3. High-energy region of kinetic energy distributions $K(E)$ for secondary electrons emitted due to impact of Ar and Kr ions, respectively. Ar ions: $E_i = 20$ eV; Kr ions: $E_i = 50$ eV. Theoretical maximum secondary electron kinetic energies indicated by arrows.

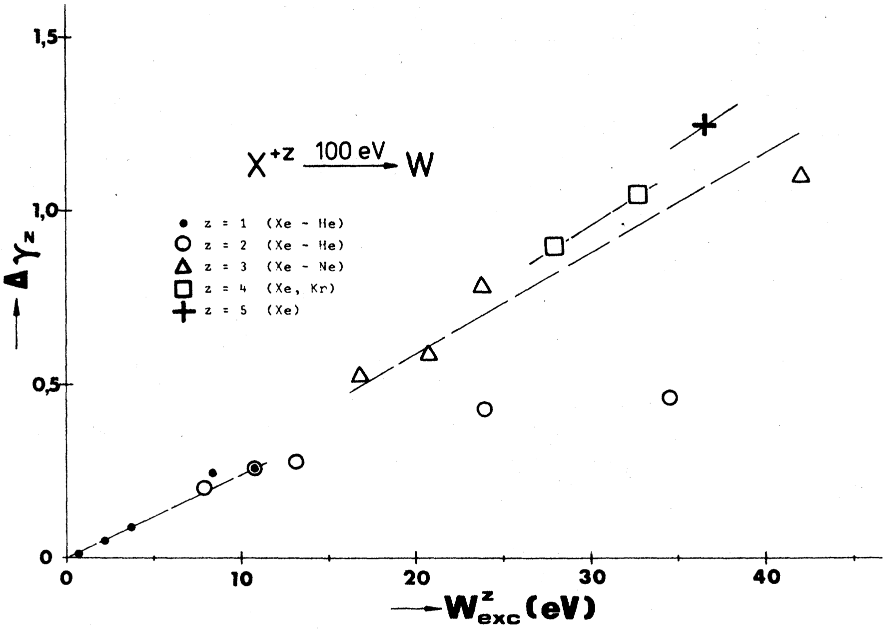

FIG. 4. Empirical relation for the dependence of secondary electron yield contribution $\Delta\gamma_z$ on excess potential energy W^z_{exc}. $\Delta\gamma_z$ data derived from γ_z data given in Table I.

fluence of metastable admixtures is seen in Fig. 3. For comparison, the high-energy regions of $K(E)$ for the doubly charged ground-state ions have been added. All pertinent E_{max} values are indicated by arrows. Similar behavior for Xe$^+$ ion beams with metastable ion admixtures has been already demonstrated.[1]

The major part of metastable doubly charged ions cannot be found from inspection of the high energy part of $K(E)$ since Table II indicates that for these metastable states relation (4) is not met.

For γ_1 values the following expression has been derived[11]:

$$\gamma_1 \simeq (\text{const})(0.8\, W_{0,i} - 2W_\phi)\,. \qquad (6)$$

For impact of 100-eV singly charged ground-state noble-gas ions on W we found (6) to hold remarkably well. If we assume that multiply charged ions are neutralized in several steps, (6) may be generalized to

$$\Delta\gamma_z \equiv \gamma_z - \gamma_{z-1} \simeq (\text{const})(0.8 W_{z-1,i} - 2W_\phi)\,, \qquad (7)$$

where $\Delta\gamma_z$ is the contribution to γ_z due to the first neutralization step. By plotting all $\Delta\gamma_z$ values as derived from Table I versus the respective "excess potential" energies

$$W^z_{exc} \equiv 0.8 W_{z-1,i} - 2W_\phi \qquad (8)$$

we have composed Fig. 4. For a fixed z, all $\Delta\gamma_z$ values are situated approximately on a straight line through the origin. Apparently, Eq. (7) may be suitable for empirical determination of γ_z. There are some significant deviations, especially for the light doubly charged ions, which, however, can be explained by a more detailed discussion of the respective ion-energy-level schemes. We note that another empirical relation[12] for γ_z differs from (7) essentially.

For metastable ions $X^{+z,m}$ we assume that the contribution to $\gamma_{z,m}$ due to the additional excitation energy $W_{z,m}$ can be given by

$$\Delta\gamma_{z,m} \equiv \gamma_{z,m} - \gamma_z \simeq (\text{const})(0.8 W_{z,m} - W_\phi)\,, \qquad (9)$$

i.e., $\Delta\gamma_{z,m}$ and thus also $\gamma_{z,m}$ can be obtained by extrapolation along the respective $\Delta\gamma_{z+1}$ line in Fig. 4 towards the value

$$W^{z,m}_{exc} \equiv 0.8 W_{z,m} - W_\phi\,. \qquad (10)$$

This procedure is justified because an ion $X^{+(z+1)}$

TABLE III. Metastable fractions in beams of singly and doubly charged noble-gas ions (cf. Figs. 1 and 2). f_{min} is the evaluation with $\gamma_{z,m} \simeq \gamma_{z+1}$; f is the evaluation with extrapolated $\gamma_{z,m}$.

Ion species	E_{IQ} (eV)	f_{min}	f
Ar$^+$	50	3.8%	4.5%
Kr$^+$	40	1.8%	2.4%
Xe$^+$	37	1.5%	2.1%
Ar^{+2}	80	1.8%	3.8%
Kr^{+2}	75	2.4%	4.2%
Xe^{+2}	60	2.5%	5.0%

may undergo resonance neutralization into excited states X^{+z*} with excitation energies that are comparable to $W_{z,m}$.

For the data shown in Figs. 1 and 2 the maximum metastable fractions f have been evaluated (a) in first approximation by assumption of $\gamma_{z,m} \simeq \gamma_{z+1}$, this procedure essentially yielding lower limits for f, i.e., $f_{\min}$, and (b) in second approximation by use of relations (9) and (10). The respective results are shown in Table III.

V. CONCLUSIONS

We have determined metastable fraction of singly and doubly charged noble-gas ion beams, respectively, by investigation of the secondary electron yield resulting from the impact of these ion beams on a clean polycrystalline tungsten surface. The metastable fractions were obtained via extrapolation for the electron yield of metastable ions from the yield measured for the respective ground-state ions. We found that the method can be applied consistently for ion impact energies up to several hundred eV. It appears that metastable fractions can be obtained accurately within 20%.

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