

Measurement of K x-ray emission from Mo, Cd, and Sn stimulated by 59.54-keV photons

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Accurate measurements, within 1.6%, of the cross section of K radiation emission of Mo, Cd, and Sn at 59.54-keV photon energy were carried out. The results are compared with those of other experiments, and they confirm the great reliability of the combination of up-to-date tabulations of K fluorescence yields and photoeffect cross-section values calculated by relativistic quantum-mechanics models.

I. INTRODUCTION

Quantitative knowledge of the emission of characteristic K radiation is still of great interest for both fundamental and applied physics. Accurate experimental values offer an appropriate means for checking the validity of the assumptions included in the different formulations of the atomic model that make it possible to evaluate important atomic parameters such as the cross section τ_k for vacancy production in the K level or the K fluorescent yield ω_k , with uncertainties in the range of some percents. In the field of applied physics, reliable values of the K emission cross section $\omega_k\tau_k$ are of great interest in elemental x-ray fluorescence analysis and, as regards the photoelectric cross section τ_k , in the aspects concerned with the contribution of the K atomic level to the attenuation of photon radiation.

Values of the K fluorescence yield ω_k were calculated in the range of atomic number $10 \leq Z \leq 70$ by Kostroun, Chen, and Crasemann [1] by combining the probabilities of radiationless transitions to a $1s$ level vacancy calculated from nonrelativistic wave functions with the radiative transition probabilities given by Scofield [2]. Chen, Crasemann, and Mark [3] used wave functions obtained from Dirac-Hartree-Slater equations to perform relativistic calculations of the same radiationless decay and combined them with Scofield's relativistic results of radiative transition probabilities (Ref. [4]) to list the ω_k values of 25 elements in the $18 \leq Z \leq 96$ range. Bambynek *et al.* [5] presented a review of selected most reliable experimental ω_k data and looked for an appropriate interpolating function, the ω_k values of which were given and compared with the theoretical ones in the $13 \leq Z \leq 92$ interval. Krause [6] worked up an ω_k tabulation in the $5 \leq Z \leq 110$ interval by using all the pertinent theoretical and experimental data on the parameters contributing to the K fluorescence yield. In a recent review Hubbell [7] calculated up-to-date fitted ω_k values in the $1 \leq Z \leq 100$ range.

An exhaustive list of researchers who worked out calculations on photoeffect cross sections can be found in a paper by Hubbell and Veigele [8]. Detailed calculations of the photoeffect cross section of each shell of the atom in the photon energy interval $10 \text{ keV} \leq h\nu \leq 3 \text{ MeV}$ for

14 elements having atomic number Z included between 13 and 92 were carried out by Schmickley and Pratt [9]. Rakavy and Ron [10] performed the calculations for five elements in the same Z interval and $1 \text{ keV} \leq h\nu \leq 2 \text{ MeV}$ but using different approaches to the atomic screening. Scofield [11] presented the results of accurate relativistic calculations of the photoeffect cross section of each atomic shell in the ranges $1 \leq Z \leq 101$ and $1 \text{ keV} \leq h\nu \leq 1.5 \text{ MeV}$ worked out with the wave function of Hartree-Slater but he also supplied a tabulation of the factors which permit conversion to the values obtainable from the Hartree-Fock model in the $1 \leq Z \leq 54$ interval. Storm and Israel [12] used the program of Brysk and Zerby [13], but with the experimental binding energies, the data of Schmickley and Pratt [9] and of Rakavy and Ron [10] and the formula of Gavrilin-Pratt [14] for preparing a tabulation of the total atomic photoeffect cross sections in the intervals $1 \leq Z \leq 100$ and $1 \text{ keV} \leq h\nu \leq 100 \text{ MeV}$. A broad and detailed review of the theoretical understanding of the photoelectric effect above 10 keV, including a comparison of results obtained by different calculation models and by experiments, is given by Pratt, Ron, and Tseng [15].

Values of the K photoeffect cross section τ_k were measured by Arora, Allawadhi, and Sood [16] for 14 elements of the atomic number interval $26 \leq Z \leq 53$ at 59.54-keV photon energy by means of a NaI(Tl) spectrometer. Garg *et al.* [17] carried out measurements of $K\alpha$ radiation emission cross sections of ten elements at 10 photon energies in the ranges $20 \leq Z \leq 56$ and $5.96 \text{ keV} \leq h\nu \leq 59.54 \text{ keV}$, respectively, by means of a Si(Li) spectrometer. Al-Nasr *et al.* [18] measured the same quantity at 59.54-keV photon energy for eight elements in the $42 \leq Z \leq 57$ range with a Ge detector.

The high performances of spectrometers using planar HPGe detectors, in terms of both resolution and efficiency, offer the possibility of experimental $\omega_k\tau_k$ evaluations having uncertainties comparable to those of the theoretical values which stem from assumptions inherent in the various models, computational errors due to rounding and limitations of arithmetic and logical algorithms in the computer. The present paper aims to achieve this purpose for the K emission cross section $\omega_k\tau_k$ of Mo, Cd, and Sn stimulated by the 59.54-keV photons emitted by a ^{241}Am source.

II. PRINCIPLE AND METHODOLOGY OF THE MEASUREMENTS

The arrangement by which the experiment was carried out is quite similar to the one by which the measurements of the atomic cross section for elastic scattering were taken [19]. The detector and the source were endowed with multivane collimators and the detector sees a region of the target foil illuminated by the whole source under specular geometry. Detector, foil, and source were assembled on a goniometric irradiation bench the reference point O of which is the intersection of source and detector collimator axes; it also belongs to the front surface of the target foil T . The goniometric bench preserves the specular geometry shown in Fig. 1 over the full range of the angles of reflection γ extending from 20° to 90° . The dimensions of the target area seen by the detector are very small in comparison with the distances k and h of O from the source surface and from the vane closest to the detector which are practically equal to one another. Furthermore, the thickness L of the target foil is much smaller than k and h . Under these conditions it is possible to show that if φ is the incident photon flux density at O and $\sigma' = d\sigma_j/d\Omega$ the differential cross section of the interaction process j between photon and atom giving rise to the emission of δ -type photons of linear attenuation coefficient μ_δ in the target material, whereas μ_i is the linear attenuation coefficient of the incident photons in the same material, when the attenuation of the air interposed between target and detector is neglected, the photon flux $\Phi_{\mu\delta}$ at the detector surface is given by

$$\Phi_{\mu\delta} = 2\varphi\Omega h k^2 I n \sigma'_j \frac{1 - \exp[-(\mu_i + \mu_\delta)L/\cos\gamma]}{\mu_i + \mu_\delta},$$

where n is the volume density of atoms in the target, Ω is the solid angle under which O sees the vane aperture closest to the detector, and I is a geometric factor which depends only on h , k , γ , r , the latter being the radius of

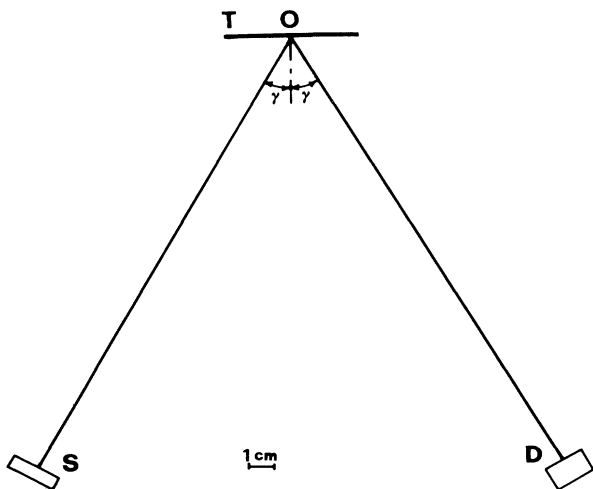


FIG. 1. Schematic layout of the experimental arrangement: S, source; D, detector; T, target foil; O, reference point; γ , reflection angle.

the circle described by the intersection of the detector collimator acceptance cone and the plane through O perpendicular to the axis of the collimator itself [19]. The attenuation of the air interposed between target and detector and having thickness L_a and linear attenuation $\mu_{a\delta}$ lowers the flux to $\Phi_\delta = \Phi_{\mu\delta} \exp(-\mu_{a\delta}L_a)$ and a further reduction is caused by the detector efficiency $\epsilon < 1$. The efficiency of a high-purity Ge planar detector of thickness L_G , with dead layer of thickness L_D and a Be window of thickness L_B for the photons of type δ , can be written as

$$\epsilon_\delta = \exp[-(\mu_{B\delta}L_B + \mu_{G\delta}L_D)][1 - \exp(-\mu_{G\delta}L_G)],$$

where $\mu_{G\delta}$ and $\mu_{B\delta}$ are the linear attenuation coefficients of Ge and Be, respectively. Then the counting rate of the spectrometer for δ -type photons is

$$N_\delta = 2\varphi\Omega h k^2 I n \sigma'_j \epsilon_\delta [\exp(-\mu_{a\delta}L_a) \times \frac{1 - \exp[-(\mu_i + \mu_\delta)L/\cos\gamma]}{\mu_i + \mu_\delta}]. \quad (1)$$

If ρ is the density of the target material, by writing

$$\mathcal{A}_\delta = \rho \frac{1 - \exp[-(\mu_i + \mu_\delta)L/\cos\gamma]}{\mu_i + \mu_\delta} = \frac{\rho f}{\mu_i + \mu_\delta}, \quad (2)$$

$$\mathcal{B}_\delta = \epsilon_\delta \exp(-\mu_{a\delta}L_a),$$

expression (1) can be written

$$N_\delta = 2\varphi\Omega h k^2 I (n/\rho) \sigma'_j \mathcal{A}_\delta \mathcal{B}_\delta. \quad (3)$$

It is worth noting the limit expressions

$$\mathcal{A}_\delta \approx \frac{\rho L}{\cos\gamma}, \quad \mathcal{A}_\delta \approx \frac{\rho}{\mu_i + \mu_\delta}, \quad (4)$$

which \mathcal{A}_δ assumes when $(\mu_i + \mu_\delta)L/\cos\gamma$ approaches zero or becomes much greater than unity.

The radiative transitions following the creation of a vacancy in the K atomic level give rise to the emission of the groups α and β of radiation, the components of each one of which are so close to one another in energy that the use of only one attenuation coefficient for each of the two groups is justified. Therefore the index δ in the expressions (2)–(4) must be replaced by α and β and if $R = p_\beta/p_\alpha$ is the ratio between the probabilities of β and α emission and $(n/\rho)_x$ the mass density of atoms in the target under study, the total α and β counting rate is given by

$$N_x = 2\varphi\Omega h k^2 I (n/\rho)_x \omega_k \tau'_k (\mathcal{A}_\alpha \mathcal{B}_\alpha + R \mathcal{A}_\beta \mathcal{B}_\beta) / (1 + R),$$

keeping in mind that the K emission differential cross section is $\sigma' = \omega_k \tau'_k = \omega_k \tau_k / 4\pi$. Likewise, when the target consists of a foil of Be whose mass density of atoms is $(n/\rho)_{\text{Be}}$ and whose atomic differential cross section for incoherent scattering is $\sigma'_j = \sigma'_{\text{KN}} S_{\text{Be}}$, where σ'_{KN} is the Klein-Nishina differential cross section and S_{Be} the incoherent scattering function of Be, the counting rate due to incoherent photons is

$$N_{\text{Be}} = 2\varphi\Omega h k^2 I (n/\rho)_{\text{Be}} \sigma'_{\text{KN}} S_{\text{Be}} \mathcal{A}_{\text{Be}} \mathcal{B}_{\text{Be}}.$$

TABLE I. Properties of the target foils. P , purity; $\mathcal{L}=\rho L$, mass thickness; $s_{\mathcal{L}}$, standard deviation of the mass thickness over the whole measured area.

Element	P (%)	\mathcal{L} (mg/cm ²)	$s_{\mathcal{L}}$ (mg/cm ²)	$(f_{\alpha})_{\pi/4}$	$(f_{\beta})_{\pi/4}$	$(f_c)_{\pi/4}$
Be	99.8	103.30	0.26			0.0427
Mo	99.9	107.30	0.41	0.968	0.931	
Mo	99.9	238.27	0.32	1.000	0.997	
Cd	99.99	203.03	0.47	0.995	0.986	
Sn	99.99	94.38	0.37	0.906	0.855	
Sn	99.99	174.52	0.67	0.987	0.972	

Therefore $\omega_k \tau_k$ can be written as a function of σ'_{KN} ,

$$\omega_k \tau_k = 4\pi \sigma'_{KN} S_{Be} \frac{M_x}{M_{Be}} \frac{N_x}{N_{Be}} \frac{(1+R)\mathcal{A}_{Be}\mathcal{B}_{Be}}{\mathcal{A}_{\alpha}\mathcal{B}_{\alpha} + R\mathcal{A}_{\beta}\mathcal{B}_{\beta}},$$

where M_x is the atomic mass of the element, the photo-stimulated K emission of which is measured, and M_{Be} the atomic mass of Be.

The experimental methodology is the same as described elsewhere [19]. The properties of the target foils used in this experiment are listed in Table I which also includes the values of $(f_{\delta})_{\pi/4} = 1 - \exp[-(\mu_i + \mu_{\delta})L\sqrt{2}]$ pertinent to the α and β groups of the characteristic K radiation and to the incoherently scattered Compton radiation, having μ_{α} , μ_{β} , and μ_c attenuation coefficients in the target material, respectively.

Keeping in mind the relations (4) and data in Table I, it is clear that most of the uncertainty regarding \mathcal{A}_{Be} comes from the standard deviation of the target thickness L while the uncertainties regarding \mathcal{A}_{α} and \mathcal{A}_{β} are mainly caused by the uncertainties of the sums $\mu_i + \mu_{\alpha}$ and $\mu_i + \mu_{\beta}$ where the heaviest addenda are μ_{α} and μ_{β} . Therefore μ_{α} and μ_{β} were measured directly.

The μ_{α} and μ_{β} measurements were carried out with the same geometric arrangement of the experiment by only inserting an additional diaphragm in proximity of the target foil. This foil was the source of the characteristic α and β radiation while another foil of the same material placed between the added diaphragm and the multivane diaphragm of the detector behaved as attenuator. The task of the added diaphragm was to assure that the attenuator is reached by the same radiation as seen by the detector in the absence of the attenuator itself. The re-

sults are summarized in Table II where the standard deviations are those arising from the uncertainties of the counting rate and attenuator thicknesses.

III. TREATMENT OF THE EXPERIMENTAL DATA

The treatment of the measurement carried out on the Be target foil strictly follows the procedure described in a previous paper [19]. In the treatment of the K counting due attention was paid to its composite structure. After subtracting the normalized background, the area of the spectral distribution, an example of which is given in Fig. 2, was evaluated in the energy interval from about 5 keV above the highest limit of the β radiation group to about 5 keV below the lowest limit of the β escape peak of the α radiation group. In order to check the results thus obtained, measurements of the α and β peaks alone, with their tails included, were performed and the correction of Axel [20] was applied. Comparison of the values given by the two procedures shows they differ by only a few tenths of a percent. The uncertainty of the net area was calculated by summing in quadrature the stochastic variabilities of the counting rates of the spectral distributions

TABLE II. Attenuation coefficients of K radiation in the target foils: P , purity; $(\mu/\rho)_{\alpha}$, average mass attenuation coefficient of α radiation group; $(\mu/\rho)_{\beta}$, average mass attenuation coefficient of β radiation group; s_{α} , standard deviation of $(\mu/\rho)_{\alpha}$; s_{β} , standard deviation of $(\mu/\rho)_{\beta}$.

Element	P (%)	$(\mu/\rho)_{\alpha}$ (cm ² /g)	s_{α} (cm ² /g)	$(\mu/\rho)_{\beta}$ (cm ² /g)	s_{β} (cm ² /g)
Mo	99.9	18.44	0.065	13.45	0.107
Cd	99.99	12.64	0.039	9.01	0.045
Sn	99.99	11.21	0.069	7.95	0.051

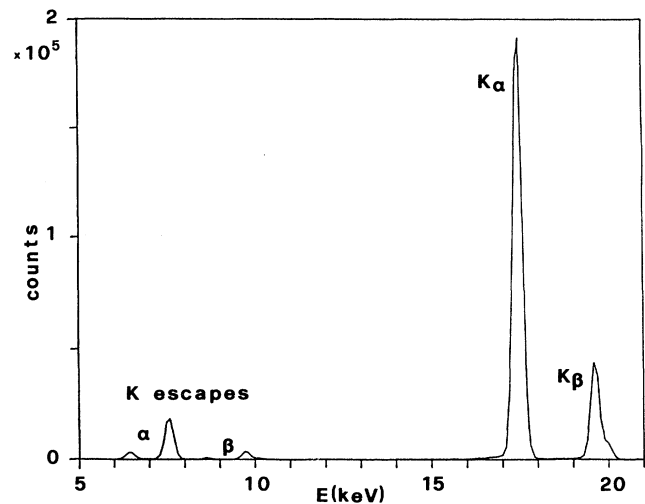


FIG. 2. Experimental spectral distribution of K radiation and of its escape peaks for a Mo target.

TABLE III. Emission cross sections of K characteristic x-rays. $s_{\omega\tau}$, absolute standard deviation of our value of $\omega_k\tau_k$; $s'_{\omega\tau}$, relative standard deviation of our value of $\omega_k\tau_k$.

Element	Z	$\omega_k\tau_k$ (b)	$s_{\omega\tau}$ (b)	$s'_{\omega\tau}$ (%)	$\omega_k\tau_k$ (b) Refs. [7,11]
		Present work			
Mo	42	437	7.0	1.6	430.8
Cd	48	773	12	1.6	776.0
Sn	50	918	14	1.5	918.5

with and without K radiation present in both the energy intervals of K radiation measurement and of normalization. So that the effect of a possible small variability in the background spectral distribution would not be neglected a conservative term equal to 0.6% of the net area was added in quadrature to the previous result.

Special attention was paid to the spurious signals present in the energy interval covered by the K radiation counting. In fact, the K photons reaching the detector owing to a photoelectric interaction in an elementary target volume are increased by the K photons produced elsewhere in the target and elastically scattered in such elementary volume having, therefore, the same energy as the former. Moreover, such outer K photons can interact incoherently in the elementary volume and they add themselves to the tail of the K peaks. This latter type of event has a negligible frequency with respect to the former for medium- and high- Z atoms and consequently a rather simple analytical model can be worked out for the calculation of the enhancement ratio. Such a ratio is appreciable whenever the irradiated area is not negligible and in the present experiment it ranges from about 1.05 to 1.08 depending on the foils with a relative standard deviation, such as is found under the calculation conditions, which, in all cases, was below 1%.

As values of the ratio $R = p_\beta/p_\alpha$, we used those calculated by Scofield [4,21], the validity of which has already been tested in the Z region of interest by Casnati *et al.* [22]. However, keeping the review of Khan and Karimi [23] in mind a standard deviation of 1% was attributed to such values. The attenuation coefficient required by the expression of \mathcal{A}_α , \mathcal{A}_β , \mathcal{B}_α , and \mathcal{B}_β were taken from four independent sources: (i) the tabulation of McMaster *et al.* [24], (ii) the tabulation of Veigele [25], (iii) the calculations of Storm and Israel [12], (iv) the calculations of Scofield [11] combined with those of Hubbell *et al.* [26]. Such attenuation coefficients enter \mathcal{A}_α and \mathcal{A}_β only for

the incident photons, and without the coherent term excluded for reasons of irradiation geometry, whereas the experimental values given in Table II were used for the K emerging photons. The correlations existing between the \mathcal{A}_α and \mathcal{A}_β values and between those of \mathcal{B}_α and \mathcal{B}_β for the closeness in energy of α and β radiation groups were properly taken into consideration in the calculation of the total standard deviation. The standard deviations coming from rectangular distributions having a half width equal to the 20% of the relevant thickness (Ref. [27]) were ascribed to the thicknesses of the dead layer L_D and of the sensitive detector depth L_G . The uncertainties of the other variables appearing in \mathcal{A} and in \mathcal{B} were neglected when they were randomized by the experiment or when their relative value was lower than 10^{-5} . For the latter reason the uncertainties of the atomic mass were also assumed to be negligible.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

In order to carry out intercomparison with the present data appropriate values of both terms of the product $\omega_k\tau_k$ are needed. For instance, the values of τ_k which can be derived from the cross section τ_a of the whole atom calculated by Storm and Israel [12] or tabulated by McMaster *et al.* [24] or by Veigele [25] are inappropriate because the calculation of τ_k from the photoeffect cross section τ_a of the whole atom by means of the relation

$$\tau_k = \tau_a \frac{r-1}{r},$$

where r is the K jump, requires that τ_k/τ_a be considered independent of photon energy, whereas as Pratt, Ron, and Tseng [15] pointed out, and Scofield's calculations showed (Ref. [11]), the ratio changes with photon energy even if only of a few percent for medium- Z atoms from K energy threshold to infinity. However, such a systematic departure cannot be accepted in the present case because it is larger than the relative uncertainty of the experimental value of the whole product $\omega_k\tau_k$. Therefore the most appropriate source of theoretical τ_k is the set of values calculated by Scofield, including the renormalization factor suggested by the same author [11].

The most up-to-date tabulation of the K fluorescence yield ω_k is the one of Hubbell [7] obtained by interpolation.

Table III contains the values measured by the present experiment, together with their absolute and relative standard deviations, and the corresponding values given

TABLE IV. Intercomparison of the emission cross sections of characteristic K x rays.

Element	Z	$\omega_k\tau_k$ (b)				
		Refs. [7,11]	Present work	Ref. [16]	Ref. [17]	Ref. [18]
Mo	42	430.8	437±7.0	430±22		404.6±8.4
Cd	48	776.0	773±12			784.5±16
Sn	50	918.5	918±14	957±39	980±34	886.0±19

by the combination of Hubbell's ω_k values and Scofield's τ_k values.

Agreement between the two series of values is very satisfactory. It is worth pointing out that just as satisfactorily an agreement is obtained with the τ_k calculated with the modified Fermi-Amaldi potential by Rakavy and Ron [10] at least for the one comparable element, i.e., Sn, for which it is $\omega_k \tau_k = 926.5$ b.

The intercomparison with the results of other analogous experiments demands the preliminary conversion to the $\omega_k \tau_k$ values of the given data. In fact, the latter refer to quantities differing from, but yet strictly linked to, $\omega_k \tau_k$. Arora, Allawadhi, and Sood [16] evaluated τ_k by dividing experimental $\omega_k \tau_k$ results by the ω_k fitted values of Bambynek *et al.* [5]; the reversion, therefore, is quite immediate. The α and β fluorescence cross sections of Garg *et al.* [17] have simply been summed to one another and expressed in barns. Al-Nasr *et al.* [18] give the α fluorescence cross section which, when expressed in barns, gives $\omega_k \tau_k$ immediately by multiplying it by $1+R$, R being the above-mentioned ratio p_α/p_β for which the Scofield [4,21] values were used. Table IV presents a synthesis of the experimental values of $\omega_k \tau_k$ for Mo, Cd, and Sn, including the ones obtained in the present work and those of the mentioned authors as well as the product of Hubbell's ω_k (Ref. [7]) and Scofield's τ_k (Ref. [11]). For the present work the uncertainties shown are standard deviations while those given by each author are used for the other experiments.

Figure 3 shows the position with respect to unity of the experimental results normalized to the Hubbell-Scofield values in the $42 \leq Z \leq 50$ interval. It is evident how the dispersion around unity is small in confirmation of the great confidence which can be attributed to the up-to-date tabulations of ω_k and to the latest quantum-mechanics models of τ_k calculation at least at the photon energy and in the atomic number interval explored.

V. CONCLUSION

The small uncertainties of the K fluorescence cross-section measurements carried out in this experiment give assurance of the accuracy of the $\omega_k \tau_k$ values which can be obtained from the up-to-date tabulations in the region of intermediate atomic numbers. This result confirms the reliability of the relativistic quantum-mechanics models used in evaluating the atomic parameters of inner shells, still further emphasized by the fact that the ω_k values

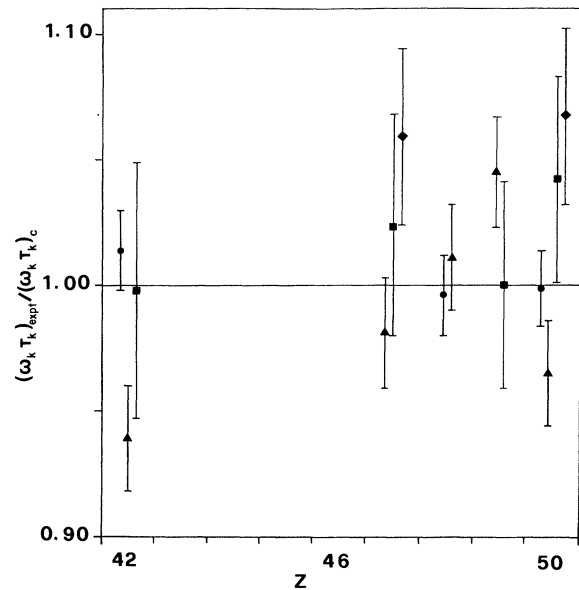


FIG. 3. Experimental cross sections of K radiation emission $(\omega_k \tau_k)_{\text{expt}}$ measured by various authors normalized to the product $(\omega_k \tau_k)_c$ of Hubbell's K fluorescence yields (Ref. [7]) and Scofield's K photoeffect cross sections (Ref. [11]). ●, present work; ▲, Al-Nasr *et al.* (Ref. [18]); ■, Arora, Allawadhi, and Sood (Ref. [16]); ◆, Garg *et al.* (Ref. [17]).

calculated by Chen, Crasemann, and Mark [3] do not differ by more than 0.3% from the interpolated ones used in the present paper. On the other hand, the accuracy of the available tabulations makes it possible to confidently employ diagnostic methodologies, where the K fluorescence cross section plays the principal role, in many important areas of human welfare and industrial activity.

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