STATISTICS OF THE EXTERNAL X-RAY PHOTOELECTRIC EFFECT OF BULK CATHODES

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Results are presented of an experimental investigation of the statistics of the x-ray photoelectric effect in metals and dielectrics in the ultrasoft region of the x-ray spectrum. An analytic expression for the probability distribution of emission acts involving different numbers of emitted electrons is derived for dielectric cathodes on the basis of widely accepted concepts regarding the x-ray photoemission mechanism. The theoretical probability distributions are in good agreement with those measured experimentally.

A characteristic feature of the external x-ray photoeffect, compared with the photoeffect in the visible region of the spectrum, is the fact that the elementary photoemission act produced by a single radiation quantum involves many electrons. The number of electrons in the "emission act," by virtue of the statistical nature of the phenomenon, does not remain constant when solitary radiation quanta are incident in succession on the photocathode, but changes from act to act. This raises the question of the probability of occurrence of an emission act with a given number of electrons. Knowledge of the statistics of the emission, i.e., of the law of the distribution of the probabilities P_n of the appearance of different numbers $n = 0, 2, \ldots$ of electrons in the elementary emission act is of exceeding importance and interest.

First, on the basis of a comparison of the experimentally measured distributions of the probabilities P_n with the theoretically calculated ones, it is possible to obtain new information on the laws governing the given phenomenon. Second, from the distribution of the probabilities it is easy to find certain characteristics of the emission, such as the quantum yield with respect to the momentum κ_{mom}, the quantum yield with respect to current κ_c, the average number of emission acts $\bar{n}$, the variance of the number of electrons in the act D, etc.; all these have to be known in connection with the solution of a number of applied-scientific problems.

Taking into account the conditions for the normalization of the probabilities

$$\sum_{n=0} P_n = 1 \tag{1}$$

the quantum yield with respect to the momenta is

$$\varkappa_{\text{mom}} = 1 - P_0 = \sum_{n=1} P_n; \tag{2}$$

The quantum yield with respect to the current is defined in this case as

$$\varkappa_c = \sum_{n=0} nP_n = \sum_{n=1} nP_n. \tag{3}$$

The average number of emission acts is

$$\bar{n} = \sum_{n=1} n \frac{P_n}{1 - P_0} = \frac{\varkappa_T}{\varkappa_{\text{mom}}}. \tag{4}$$

It should be noted that a determination of these quantities by other methods entails exceedingly laborious absolute measurements of the fluxes of x-ray quanta and of the emitted electrons. Recently, many direct methods for the experimental study of the statistics of electron emission of different types have been developed. New data were acquired on the statistics of secondary electron emission,[1-7] ion-electron emission,[8-10] electron emission due to neutral atoms[10,11] and particles,[12] and other types of electron emission.[13]

In [14] it was demonstrated that in principle it is possible to study the statistics of the x-ray photoeffect with the aid of a method based on acceleration of the emitted electrons to high energies and subsequently registering them with a small-size proportional counter with an entrance window made of thin organic film. The present paper is a continuation of [14]. We present here the results of an experimental study of the statistics of the x-ray photoeffect from metals and dielectrics in the ultrasoft region of the x-ray spectrum. The probabilities for the appearance of emission acts with different electron numbers are calculated for dielectric photocathodes. The experimentally measured distributions of P_n are compared with the theoretically calculated ones.

EXPERIMENTAL PART

The measurements were carried out in the vacuum x-ray monochromator,[15] the design of which was modified and improved somewhat to increase the contrast and the intensity of the monochromatic x-radiation. A simplified diagram of the setup is shown in Fig. 1.

The procedure for experimentally determining the distribution of the probabilities P_n consisted in the following. A narrow beam of monochromatic quanta was incident on a flat photocathode 10 and caused emission of electron "batches." A proportional electron counter 11 was placed opposite the photocathode, at a distance 3–5 mm. An 8–10-kV accelerating voltage was applied between the photocathode and the body of the counter. This voltage should ensure favorable conditions for the gathering of electrons from the photocathode into the counter and conditions for the passage of the electrons

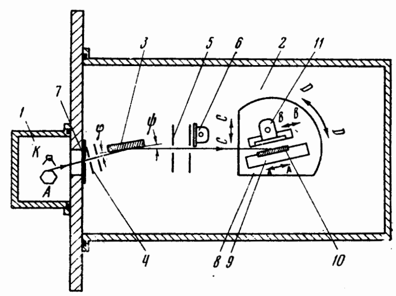

FIG. 1. Schematic diagram of an ultrasoft x-ray monochromator: 1–K–cathode, A–anode, 2–volume of monochromator, 3–rotating diffraction grating, 4, 5–immobile entrance and exit slits of monochromator, 6–counter-monitor for the measurement of the absolute number of quanta of x-radiation incident on a flat photocathode, 7–dividing slit, 8–unit for securing the photocathode and the electron counters, C–C and D–D–possible variants of motion of the given unit in the volume of the monochromator, 9–holder for photocathode 10, which can move in the direction A–A, 11–proportional electron counter, which can move in the direction B–B.

through the film of the window without noticeable energy loss. It can be shown, considering a cosinusoidal distribution of the electrons with respect to the emission angles[16] and taking their energy composition into account,[17] that when the emitting area of the photocathode is $0.1 \times 3\ mm^2$ and the area of the entrance window of the counter is $1 \times 15\ mm^2$, practically all the emitted electrons fall into the counter at voltages 8–10 kV. The electron loss due to reflection and absorption by the thin organic film of the counter (approximate thickness 1000 Å), in accordance with the published data[18, 19] and the experimentally measured transmission curve, amounts to several percent. The total electron-gathering coefficient is close to unity.

The "batches" of accelerated electrons, on falling into the counter, produce pulses with amplitude proportional to the total energy of the "batch." If strictly single-electron "batches" introduce into the counter an energy E_1 and produce a momentum distribution with average amplitude U_1, then strictly n-electron "batches" introduce into the counter an energy nE_1 and produce a momentum distribution with average amplitude nU_1. An analysis of the pulses by amplitudes was realized with the aid of the AI-100 pulse-height analyzer. Examples of the amplitude distribution of the pulses for certain cases of x-ray photoemission are shown in Fig. 2.

The experimentally measured distributions of the pulses by amplitude $X(U)$ constitute an additive superposition of amplitude distributions $X_n(U)$ for strictly n-electron emission acts with a definite statistical "weight" p'_n (where n = 1, 2, 3, ...), i.e.,

$$X(U)=\sum_{n=1} X_n(U)P_n', \quad n=1,2,3,\ldots. \tag{5}$$

The distribution of the "weights" P'_n represents a distribution of the relative probabilities of the acts of emission of a given number of electrons. The distribution $X_n(U)$ for n = 2, 3, 4, ... was calculated from the recurrence formula

$$X_n(U)=\sum_{Z=0}^{U} X_1(Z)X_{n-1}(U-Z), \tag{6}$$

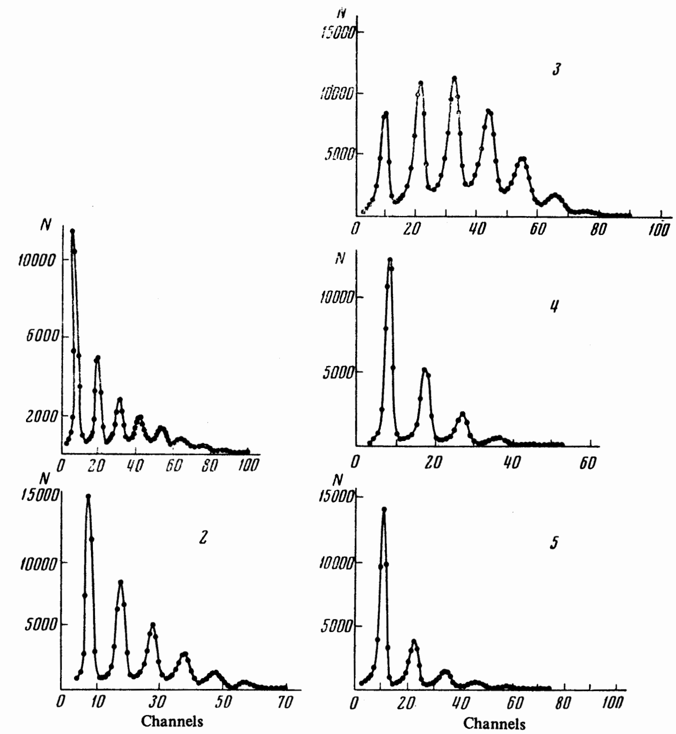

FIG. 2. Experimentally measured amplitude distributions of the pulses from the proportional counter of the electrons (N–number of counts per channel): 1–NaCl photocathode, hν = 185 eV; 2–KBr, hν = 114 eV; 3–CsCl, hν = 96.5 eV; 4–CsI, hν = 65.2 eV; 5–Au, hν = 525 eV. Glancing angle φ = 20°.

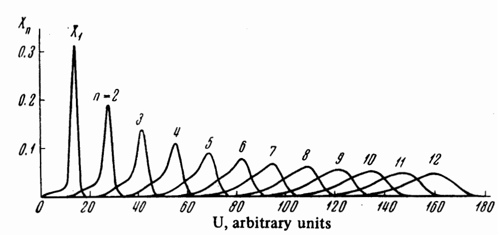

FIG. 3. Amplitude distributions of pulses from proportional counter of electrons, calculated with the aid of formula (6) for strictly n-electron emission acts (n = 2–12); X_1–experimentally obtained function of amplitude distribution of the pulses from the counter for single-electron emission acts.

where $X_1(U)$ is the experimentally measured distribution of the momenta with respect to the amplitudes for strictly single-electron emission acts, the sources of which were specially prepared light-sensitive photocathodes.

Formula (6) was used to determine, with the aid of a BESM-3 computer, the values of $X_n(U)$ for n from 2 to 14. Figure 3 shows the experimental amplitude distribution $X_1(U)$ and the values of $X_n(U)$ calculated by formula (6). Since the films of the entrance window of the electron counter failed frequently, the aforementioned calculations of $X_n(U)$ were made for each new film used.

Thus, from the experimental distribution of the pulses by their amplitudes $X(U)$ and from the distributions $X_n(U)$ calculated by solving a system of n linear equations with unknown "weights" P'_n we determined the values of P_n'. All these calculations were performed with the computer. Figure 4 shows the distribution of the probabilities P_n' of the occurring emission acts, obtained for the experimental distributions X(U) shown in Fig. 2. The distribution of P_n' was normalized to unity, i.e.,

$$\sum_{n=1} P_n' = 1.$$

It should be noted that the probability of a zero photoemission act P_0 cannot be determined from X(U) directly. The value of P_0 can be found, as was done in [11, 20], by extrapolating P_n'. Since in this case there is always a certain indeterminacy in the choice of P_0, in the present study P_0 was determined, in addition, by measuring the momentum quantum yield κ_{mom} (see (2)). The value of κ_{mom} was measured as the ratio m/m_0, where m is the number of emission acts registered by the proportional electron counter or by a secondary-electron multiplier of the open type, and m_0 is the number of x-ray quanta incident on the photocathode, which were registered by the quantum counter 6 (Fig. 1) with known efficiency. The accuracy of the determination of κ_{mom} was 10–15%.

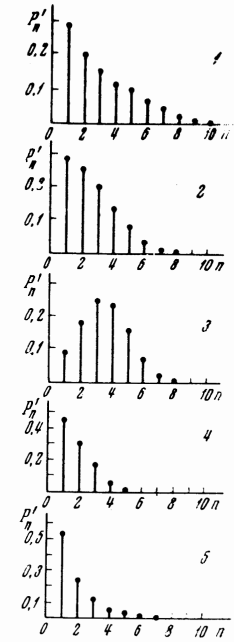

FIG. 4. Distribution functions of the probabilities P_n' for the appearance of emission acts with different numbers of electrons n for different photocathodes and different wavelengths: 1–NaCl photocathode, $h\nu$ = 185 eV, 2–KBr, $h\nu$ = 114 eV, 3–CsCl, $h\nu$ = 96.5 eV, 4–CsI, $h\nu$ = 65.2 eV, 5–Au, $h\nu$ = 525 eV. Glancing angle φ = 20°.

CALCULATION OF P_n AND COMPARISON WITH EXPERIMENT

Starting from the existing notions [21] concerning the laws governing the formation of the act of x-ray photoemission in dielectrics, we can obtain an analytic expression for the law of the distribution of the probabilities P_n. The following assumptions are made here.

1. The x-ray quanta of ultrasoft radiation incident on the flat photocathode at a glancing angle[1)] φ penetrate into the interior of the photocathode and are absorbed in a layer dx at a distance x from the surface with a probability

$$\frac{\mu}{\sin\varphi}\exp\left(-\frac{\mu x}{\sin\varphi}\right)dx,$$

where μ is the linear coefficient of attenuation of the x-rays.

2. When a quantum is absorbed in a layer dx, fast photoelectrons and Auger electrons are produced; when these electrons move in the photocathode material they excite slow secondary electrons. For the considered x-radiation wavelength region, the photoelectrons and the Auger electrons of the investigated cathodes have energies on the order of several dozen or several hundred electron-volts. At these energies, the free path of the fast x-ray electrons in the dielectric is smaller than the free path of the secondary electrons.[22] Therefore, in first approximation, it is possible to disregard the individual trajectories of the motion of the primary x-ray photo- and Auger electrons and to disregard the functions of the excitation density of the secondary electrons on their entire path of motion, assuming that the "swarm" of secondary electrons occurs at that position in the photocathode where the absorption of the x-ray quantum took place, i.e., in the layer dx at a depth x from the surface of the emitter. The number N of the secondary electrons in the "swarm" is determined by the quantum energy $h\nu$ and by a certain energy E, which must be consumed to produce one secondary electron capable of leaving the solid. The quantity E includes the kinetic energy of the emerging secondary electron and all the energy losses connected with the heating of the body, with emission, with overcoming the potential barrier on the emitter surface, and other possible types of losses. On the average, the creation of one secondary electron capable of emerging to the vacuum requires the consumption of an energy E_0 and consequently the average number of secondary electrons occurring upon absorption of an x-ray quantum $h\nu$ is

$$N_0 = h\nu/E_0.$$

It should be noted that N is a fluctuating quantity. The law of variation of N in different acts of x-ray-quantum absorption is not known. Two extreme cases of fluctuations of the number N are considered:

a) N has a Poisson variation

$$K(N) = \frac{N_0^N}{N!}e^{-N_0},$$

b) $N = N_0$ = const.

3. In accordance with the usual assumption made in different theories on secondary electron emission and photoemission,[23, 24] and also with allowance for the results of the latest investigations,[7, 25] where the Monte Carlo method was used to calculate the probability of emission of slow electrons from dielectrics, it is assumed that any electron of the "swarm" produced at a depth x in the layer dx reaches the surface of the emitter and leaves it with a probability

$$q(x) = Ae^{-\alpha x},$$

where $1/\alpha$ is the mean free path of the secondary electron and A is the probability of emergence of the electron into the vacuum in the case where it is produced at the very surface of the photocathode.

[1)]We consider the region of angles at which there is no total external reflection of the x-radiation.

Table I

P_n	NaCl ($h\nu$ = 185 eV)		KBr ($h\nu$ = 114 eV)		CsCl ($h\nu$ = 96,5 eV)		CsI ($h\nu$ = 65,2 eV)	
	experiment	calculation $n_0 = 16$ $1/\xi = 5{,}0$	experiment	calculation $n_0 = 9$ $1/\xi = 1{,}6$	experiment	calculation $n_0 = 8$ $1/\xi = 0.2$	experiment	calculation $n_0 = 5$ $1/\xi = 1{,}1$
P_0	0.6300	0.6000	0,330	0,3316	0,020	0,0215	0,370	0.3531
P_1	0,1060	0,1200	0,191	0,2060	0.086	0,0882	0,286	0,2926
P_2	0,0750	0.0620	0,170	0.1619	0,176	0,1865	0,193	0.2083
P_3	0,0575	0.0530	0.135	0,1270	0,246	0,2528	0,108	0,1073
P_4	0,0430	0.0420	0.090	0,0895	0,230	0,2322	0,034	0.0338
P_5	0,0380	0,0340	0,053	0,0520	0,147	0,1446	0,008	0.0048
P_6	0,0240	0,0270	0.020	0,0231	0,068	0,0586		
P_7	0,0160	0,0210	0,007	0,0072	0.017	0,0140		
P_8	0,0075	0,0140	0.003	0,0014	0,002	0,0015		
P_9	0,0029	0,0080						
P_{10}	0.0006	0.0044						

It should be noted that in our case by $1/\alpha$ is meant the mean effective free path of the secondary electrons, with allowance for their energy distribution inside the solid and for the dependence of the free path on the energy. The coefficient A, as shown in [7, 25], also depends on the energy of the secondary electrons, and we therefore assume that it has a certain mean effective value lying in the range from ½ to 1. To carry out the calculation within the framework of the assumptions there is no need to employ concrete mechanisms of interaction between the secondary electrons and the solid. It can be noted, however, that the assumption of an exponential variation of the probability of the emergence of the electrons from the dielectric apparently presupposes there an electron-phonon mechanism of interaction of the secondary electrons.

Thus, if it is assumed that the probability of emergence of each of the "swarm" electrons from the photocathode is $q(x) = A \exp(-\alpha x)$, then the probability of simultaneous emergence of n electrons out of the N produced in the layer dx at a depth x is determined by the binomial distribution

$$T(n) = C_N{}^n [Ae^{-\alpha x}]^n [1 - Ae^{-\alpha x}]^{N-n}.$$

On the other hand, the probability of appearance in the external emission of n electrons out of N produced as a result of absorption of an x-ray quantum in a layer dx at a distance x from the surface of the photocathode is

$$M(n) = \frac{\mu}{\sin\varphi} \exp\left(-\frac{\mu x}{\sin\varphi}\right) C_N{}^n [Ae^{-\alpha x}]^n [1 - Ae^{-\alpha x}]^{N-n}.$$

Integrating over all layers dx from 0 to ∞, we find that the probability of appearance of n electrons in the emission act can be represented in the form

$$P_n = \sum_{N=n}^{\infty} \frac{N_0{}^N}{N!} e^{-N_0} C_N{}^n \sum_{k=0}^{N-n} (-1)^k A^{k+n} C_{N-n}^k \frac{\xi}{\xi + (n+k)}, \qquad (7)$$

if N has a Poisson variation, and in the form

$$P_n = C_N{}^n \sum_{k=0}^{N-k} (-1)^k (A)^{k+n} C_{N-n}^k \frac{\xi}{\xi + (n+k)}, \qquad (8)$$

If $N = N_0 = \text{const}$. Here $\xi = \mu/\alpha \sin\varphi$.

Formulas (7) and (8) were used for a computer calculation of the probabilities P_n for different values of N_0 and ξ, and also for A = 0.5, 0.75, and 0.9; these values were then compared with the experimentally measured distributions. It turned out that the P_n calculated by formula (7) did not agree with experiment,

Table II

	NaCl ($h\nu$ = 185 eV)	KBr ($h\nu$ = 114 eV)	CsCl ($h\nu$ = 96,5 eV)	CsI ($h\nu$ = 65,2 eV)	Au ($h\nu$ = 525 eV)
$\varkappa_{mom}$	0,37	0.67	0,98	0.63	0,087
$\bar{n}$	3,1	2,6	3.5	1.8	1.9
$\varkappa_c$	1,13	1.76	3.42	1,13	0.16
D	3,98	2,24	1,99	1.18	1.53

whereas the P_n calculated by formula (8) agree well with the experimental data. As evidence for the small fluctuation of the number N we might cite the results of [26], where the values of the Fano factor were calculated for the primary ionization produced by incident electrons in Ge and Si. These factors were respectively $F_{Ge} = 0.1-0.2$ and $F_{Si} = 0.05-0.1$. Such small values of the Fano factor for solids offer evidence of the small value of the variance $D = \overline{(N - \bar{N})^2} = F\bar{N}$, i.e., it can be assumed that N is practically equal to $\bar{N} = N_0$.

Table I gives the experimental values of P_n and those theoretically calculated by means of formula (8) for photocathodes of NaCl, KBr, CsCl, CsI, and Au and for different wavelengths.

It should be noted that the agreement of the experimentally obtained distributions P_n with the theoretical ones is possible only under fully defined and unique values of N_0 and ξ, which determine the statistics of the given type of x-ray photoemission. Knowledge of the quantities N_0 and ξ makes it possible to find the important characteristics α and E_0 of x-ray emission.

Table II lists the values of κ_{mom}, κ_t, $\bar{n}$, and the variance D, obtained from the experimentally measured distributions P_n given in Table I.

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