Quantum yield of the x-ray photoelectric effect in negative electron affinity emitters

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An investigation of the external x-ray photoelectric effect in emitters with negative electron affinity (NEA) was made in the spectral range 0.05–0.25 nm. The experimental techniques and the method are described. The experimental absolute values of the quantum yield of NEA emitters are reported for normal incidence of x rays on the photocathode in the energy range 17–5 keV. These values of the quantum yield are one or two orders of magnitude higher than for the conventional x-ray photocathodes. The results of studies of the spectral and angular dependences of the photoelectric effect are reported for $GaP_{0.4}As_{0.6}$, which is one of the most efficient NEA photocathodes. A simple theoretical model of the x-ray effect in NEA emitters is put forward and an analysis is made of the experimental results. Good agreement between the experiment and theory is achieved. It is shown that such important parameters of the photoelectric effect as the diffusion length of electrons and the probability of electron emission from the emitter surface in vacuum can be determined.

Emitters with a negative electron affinity (NEA) are used increasingly in various branches of science and technology as efficient electron sources. The progress has been particularly marked in studies and practical applications of NEA emitters as secondary-emission materials and photocathodes for the visible part of the spectrum.[1] Studies of the x-ray range are lagging far behind. There are few papers on this topic.[2,3] However, the need for x-ray NEA photocathodes is very urgent because of applications in x-ray spectroscopy, microscopy, and astronomy, in diagnostics of pulsed sources of x rays, of laser plasma, etc.[2–14] In view of the specific nature of the x-ray photoeffect we can obtain new data on the physical properties of the surface layers of semiconductors, including the processes of interaction between thermalized electrons with the crystal lattice and on the energy structure of the conduction band.

We shall try to fill the gap in this subject. The study reported below was in a sense pioneering. The aim was to determine the absolute values of the quantum yield of the x-ray photoeffect in NEA emitters made of III-V compounds and their solid solutions, in a wide spectral range and for different angles of incidence on the photocathode, and to determine (from a comparison of the experimental data with the theoretical representations given below) such important characteristics of emitters as the diffusion length of electrons and the probability of the emission of an electron from a surface in vacuum. We also investigated the stability of the quantum yield with time and the homogeneity of the photoemission of surfaces of emitters in order to determine potential applications of NEA emitters in x-ray instruments.

TECHNIQUES AND EXPERIMENTAL METHOD

Our measurements were carried out using apparatus based on an x-ray diffractometer described in Ref. 15. Figure 1 shows a simplified schematic diagram of the apparatus. A sealed glass device 1 with a thin beryllium window 2 for the entry of an x-ray beam was used as a receiver chamber. Inside the sealed device a moving cassette 3 carried eight investigated samples 4. Displacement and rotation of the cassette with the samples about the central axis 5 of the device were due to the application of a field by a magnet 15. An angular scale was attached to the outer surface of the device. The device 1 was bonded to a special platform 6 which made it possible to rotate the device about the vertical axis in order to set the axis of rotation of photocathodes at right angles to the x-ray beam and also to be able to move the device along two mutually perpendicular directions in the course of scanning of the photocathode surface with an x-ray beam.

A beam of x rays from a demountable x-ray tube 7 passed through a stop 8 and reached an LiF monochromator crystal 9 with the (200) orientation, placed on the axis of a goniometer 10. A monochromatic beam reflected by lithium fluoride reached at an angle φ the photocathode 4 after passing through an entry slit 11 and a window 2, and then through the glass device 1. The emitted electrons were collected by a collector 12, which was subjected to an accelerating voltage of + 300 V. The intensity of the radiation incident on the photocathode was measured using a proportional counter 13 located on a moving stage in front of the exit window 2, which made it possible to remove the

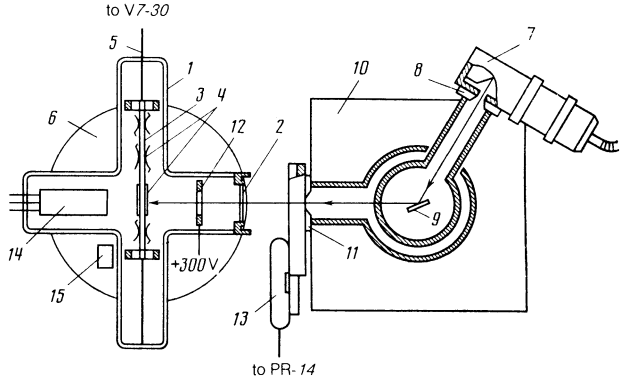

FIG. 1. Schematic diagram showing the apparatus (explanations in text).

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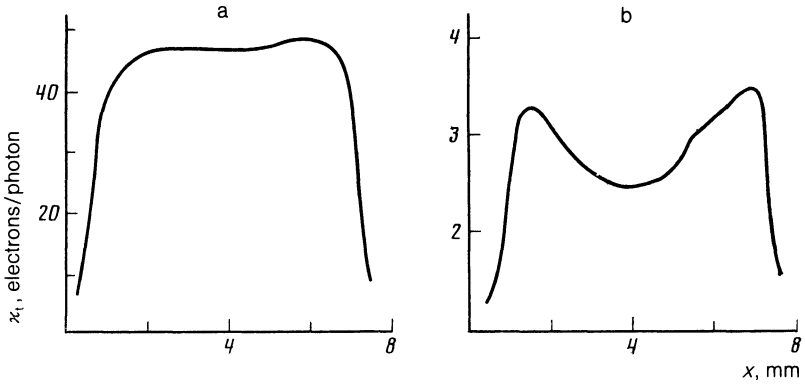

FIG. 2. Dependence of the quantum yield $\varkappa_t$ on the point of incidence of the x-ray beam x on the surface of a photocathode. The results are plotted for MoK_α radiation.

counter from the x-ray beam during measurement of the photocurrent pulse. The photocurrent was determined with a V7-30 electrometer characterized by a low-resistance input. This input was connected to the central axis 5 along which a cassette with the samples 3 could be slid. The absence of an observable charging of the photocathodes during the photocurrent measurements justified the use of a sliding contact. Measurements of the leakage resistance along the glass envelope of the device and the connecting cable exceeded the input resistance of the electrometer in its most sensitive range by more than 1000. Inside the glass device there was a source of cesium used to activate the photocathode and an electron gun 14, which made it possible to determine the secondary electron emission coefficient for various energies of the primary beam. The surfaces of the samples were cleaned by general heating in ultrahigh vacuum before activation. The glass device was screened from light and electrostatic fields by a nickel foil.

The sources of x rays were serially manufactured BSV-27 x-ray tubes with different anodes. Measurements were made using the K_α and K_β lines of V, Cr, Fe, Co, Cu, Mo, and Ag, as well as in the region of the K absorption edges of Ga and As against the continuous background of x rays emitted from a tube with a molybdenum anode. The power supplied to the tubes was selected so as to avoid higher reflection orders from the LiF monochromator. The line contrast was at least 100.

We investigated dynodes taken from various photomultipliers with NEA emitters based on polycrystalline GaP_xAs_{1-x} and GaP–Mo structures, as well as plane single-crystal $GaPo_{0.4}As_{0.6}$ photocathodes grown by the method of vapor epitaxy on GaAs single crystals. All the samples had p-type conduction and were doped with Zn to give an acceptor concentration of 10^{19} cm^{-1}. The samples were activated to the state of a negative electron affinity by cesium and oxygen.

The quantum yield $\varkappa_t$ of the x-ray photoelectric effect was measured on the basis of the electric current. Its value was deduced from the relationship $\varkappa_t = ik/eN$, where i is the photocurrent measured with the electrometer, e is the electron charge, N is the number of photons incident per second (measured with a counter), and k is an experimentally determined coefficient allowing for the absorption of x rays of a given wavelength in the air gap between the counter window and the entry window of the sealed glass device, and also for the absorption in the beryllium entry window and for the absolute efficiency of the counter. The quantum yield values quoted below will always be in absolute units of electrons/photon.

EXPERIMENTAL RESULTS

We investigated two batches of eight samples each. These batches differed from one another in the degree of activation. In the first batch the measured quantum yield was maintained for a year and then began to fall. In the second batch we found that seven samples retained their properties after five months and in the case of one sample the quantum yield fell by 20% in this period.

The uniformity of photoemission across the surface was monitored by measuring the quantum yield of the individual parts of the photocathode scanned with a narrow (0.5×0.5 mm) x-ray beam which was guided along the surface of a sample along two mutually perpendicular directions. Figure 2 shows, by way of example, typical dependence of the quantum yield $\varkappa_t$ on the position x of the x-ray beam on the surfaces of single-crystal (a) and polycrystalline (b) photocathodes in the case of normal incidence of the x-ray beam. The nonuniformity along the single-crystal surface was less

TABLE I. Experimental values of quantum yield $\varkappa_t$ (electrons/photon) of the investigated photocathodes ($\varphi = 90°$).

Radiation wavelength, nm	Sample No.							
	1	2	3	4	5	6	7	8
Mo K_α, λ=0,07	49	48	4.5	4.3	3.3	4.7	1.1	2.1
V K_α, λ=0,25	60	58	–	–	5.7	6.6	2.3	3.4

Note. Columns 1 and 2 give the values for single-crystal $GaP_{0.4}As_{0.6}$ photocathodes; columns 3–6 represent polycrystalline $GaP_{0.4}As_{0.6}$ photocathodes; columns 7 and 8 represent polycrystalline GaP photocathodes.

TABLE II.Experimental values of quantum yield $\varkappa_{t.e}$ for a single-crystal photocathode, measured at different wavelengths λ, compared with calculated values $\varkappa_{t.c}$.

	Radiation wavelength λ, nm						
	MoK_α, 0.07	CuK_β, 0.144	CuK_α, 0.154	FeK_β, 0.175	FeK_α, 0.193	VK_β, 0.228	VK_α, 0.25
$\varkappa_{t.e}$, electrons/photon	49	28	33	41	44	54	60
$\varkappa_{t.c}$, electrons/photon	60	27	33	39	43	53	58
μ, cm^{-1}	268	239	317	454	597	941	1220

than 5%. The variation of the quantum yield of the polycrystalline sample in the course of scanning with the x-ray beam was due to the angular dependence $\varkappa_t(\varphi)$ resulting from the curvature of the dynode surfaces.

The values of the quantum field obtained for the second batch of samples, measured at the wavelengths of $\lambda = 0.25$ and 0.07 nm in the case of normal incidence of the x-ray beam on the photocathode surface, are listed in Table I. The quantum yield for the first batch of samples was somewhat lower. It is clear from Table I that the quantum yield of polycrystalline samples was considerably less than the quantum yield of single crystals. In the case of $GaP_{0.4}As_{0.6}$ single crystals the quantum yield reached 60 electrons/photon, which was one or two orders of magnitude higher than the values of the quantum yield for conventional x-ray photocathodes.[14,16,17] Single-crystal $GaP_{0.4}As_{0.6}$ photocathodes were selected, as the most efficient among the investigated samples, for a detailed study of the angular and spectral dependences of the quantum yield.

Table II gives the values of the quantum yield obtained for normal incidence of the x-ray beam on a $GaP_{0.4}As_{0.6}$ single crystal, determined in the wavelength range 0.07–0.25 nm which included the K absorption edge of As ($\lambda = 0.104$ nm) and Ga ($\lambda = 0.119$ nm). A monotonic rise of the quantum yield on increase in the wavelength was observed beyond the absorption edges of Ga and As.

Table III gives the results of a determination of the angular dependence on the quantum yield $\varkappa_t(\varphi)$ in the range of glancing angles from 7 to 90°, obtained for one of the $GaP_{0.4}As_{0.6}$ photocathodes at wavelengths of $\lambda = 0.25$ and 0.14 nm. It is clear from Table III that at $\lambda = 0.25$ nm the quantum yield increased twofold when the glancing angle was reduced from 90 to 7°, whereas at $\lambda = 0.14$ nm the quantum yield increased sixfold in the same range of angles.

DISCUSSION OF RESULTS

The results were analyzed using the theory of the x-ray photoelectric effect developed in Refs. 2 and 18 and a diffusion model of the transport of electrons toward the surface.[1,19] Before applying the diffusion model in the calculation of the characteristics of an NEA emitter, it was necessary to select a suitable excitation function $G(x)$ describing the distribution of the thermalized electrons and holes, generated as a result of absorption of x rays.

If a monochromatic x-ray beam with photons of energy $h\nu$ is incident on a semiinfinite photocathode at an angle φ relative to the plane surface, then for glancing angles φ higher than the critical angle of total external reflection, the probability of absorption of a photon in a layer dx at a depth of x from the surface is

$$F(x)\,dx = \frac{\mu}{\sin\varphi}\exp\left(-\frac{\mu x}{\sin\varphi}\right)dx, \qquad (1)$$

where μ is the linear absorption coefficient of the photocathode material. The absorption of photons by atoms in this material results in creation of electrons with the following different energies.

1. The K, L, M, etc. photoelectrons knocked out from the various atomic shells with energies $h\nu - E_{ij}$, where E_{ij} is the binding energy of an electron of the ith atom in the jth shell.

2. The Auger electrons of various kinds, corresponding to nonradiative transitions of an atom from an excited to the ground state with energies $E_{ij} - \Delta E_{ls}$, where ΔE_{ls} is the energy of the corresponding nonradiative transitions in the ith atom when the jth shell is excited, creating an Auger electron of a given kind.

3. The photoelectrons formed as a result of absorption of fluorescence photons $h\nu_{ij}$ created by radiative transitions in the course of deionization of the jth shell of atoms of the ith kind that have absorbed a photon $h\nu$ in the jth shell. The energy of these photoelectrons is $h\nu_{ij} - E_{ls}$, where E_{ls} is the binding energy of electrons in the absorbing atom.

We shall call all these electrons primary, by analogy with the terminology used in secondary electron emission. The sum of the energies of the primary electrons is equal to the energy of the absorbed photon. The range of primary electrons expressed in centimeters is given by the relation[20,21]

$$l_j = kAE_j^n/\rho Z, \quad n = 1-2,$$

TABLE III. Experimental values of quantum yield $\varkappa_t$ (electrons/photon) measured for different glancing angles φ.

λ, nm	φ°											
	7	11	14	16	18	21	25	29	36	43	54	90
0.144	168	121	96.3	91.3	–	69.4	62.2	56	49.3	43.1	34.7	28
0.25	121.2	123.6	118.8	–	106.8	100.8	96	91.2	82.8	74.4	69.6	60

where ρ is the density of the photocathode material, Z is the atomic number, A is the atomic weight, $k = 5\times 10^{-16}$, and E_j is the energy of an electron in kiloelectron-volts. In the investigated part of the spectrum for semiconductor materials used as the base for NEA emitters the maximum range l of a fast electron amounts to a few tenths or hundredths of a micron. In the course of motion in the medium the primary electrons with relatively high energies excite in a path l a cascade of secondary "hot" electrons and the bulk of these secondary electrons is excited in a distance much shorter than l. If we assume that the creation of one secondary electron requires on the average an energy ε, it follows that the absorption of one photon creates $n_0 = h\nu/\varepsilon$ secondary electrons. The energy ε allows for the energy losses in radiative transitions in an atom, for the energy used to heat the solid, and for the energy carried away by fast and slow electrons outside the solid. As a result of repeated energy dissipation these excited electrons may reach an equilibrium state (they may be thermalized) at metastable levels at minima of the conduction band where their lifetime is limited only by the process of their recombination with holes.

If the diffusion length of thermalized electrons L is considerably greater than the range of primary electrons l and the thermalization length of "hot" secondary electrons L_t, then in the first approximation we may assume that the thermalized electrons are generated at the point where a photon is absorbed. Then, the generation function $G(x)$ is governed by the probability of absorption of an x-ray photon $F(x)dx$:

$$G(x)=n_0\frac{\mu}{\sin\varphi}\exp\left(-\frac{\mu x}{\sin\varphi}\right)dx. \qquad (2)$$

Then, strictly speaking, we can apply the diffusion model only if the depth of absorption of x rays exceeds the values of l and L_t. We can determine the range of validity of these assumptions by considering the mechanism of relaxation of the energy of "hot" electrons.

One of these mechanisms in semiconductor materials is the scattering by polar optical phonons. Using the results of Ref. 22, we can show that the thermalization length of "hot" electrons of energy 0.3 eV is less than 0.2 μm. However, the bulk of the energy of "hot" electrons is lost over distances less than 0.1 μm, as indicated by the results of determination of the depth of emission of electrons by secondary electron emission from efficient emitters with a low positive electron affinity.[21,23] An analysis of other energy relaxation mechanisms in heavily doped p-type semiconductors (scattering by heavy holes[24] and by coupled plasmon–phonon oscillations[25,26]) gives even smaller values of L_t.

The depth of absorption of x rays incident normally on a photocathode amounts, in the investigated spectral range and for the investigated objects, to a few microns or tens of microns, which is considerably greater than l or L_t. Determination of the quantum yield of NEA emitters based on III-V compounds in the optical part of the spectrum[1] gives diffusion lengths of thermalized electrons L amounting to several microns, which is again much greater than l or L_t. Therefore, to lowest order, we can describe the x-ray photoeffect using the assumptions made above. It should be pointed out that at small glancing angles the above assumptions about the relationships between l, L_t, L, and $1/\mu$ are no longer justified and the characteristics of x-ray NEA emitters should be determined using, for example, the approach developed in Ref. 16.

It follows from the diffusion theory of motion of electrons in a solid that a thermalized electron reaches the surface from a depth x where it is generated with a probability $\exp(-x/L)$ and escapes into vacuum with a probability $A\exp(-x/L)$, where A is a constant with values in the range $0\leqslant A\leqslant 1$. Therefore, the number of electrons escaping into vacuum from a layer dx at a depth x is (per incident photon)

$$dn=n_0\frac{\mu}{\sin\varphi}\exp\left(-\frac{\mu x}{\sin\varphi}\right)A\exp\left(-\frac{x}{L}\right)dx. \qquad (3)$$

The quantum yield in the case of absorption of x rays throughout the thickness of the photocathode can be found by integrating Eq. (3) with respect to x from zero to infinity:

$$\varkappa_t=\int_{x=0}^{x=\infty}dn=An_0\left(1+\frac{\sin\varphi}{L\mu}\right)^{-1}. \qquad (4)$$

Using Eq. (4), we can determine the parameters L and A by altering the values of the quantum yield $\varkappa_{t1}$ and $\varkappa_{t2}$ for any pair of radiations with photon energies $h\nu_1$ and $h\nu_2$ and a given glancing angle φ:

$$L=\frac{\sin\varphi}{k_1-k_2}\left(\frac{k_2}{\mu_1}-\frac{k_1}{\mu_2}\right),\quad k_1=\frac{\varkappa_{t2}}{\varkappa_{t1}},\quad k_2=\frac{h\nu_2}{h\nu_1}, \qquad (5)$$

$$A=\frac{\varkappa_t\varepsilon}{h\nu}\left(1+\frac{\sin\varphi}{L\mu}\right). \qquad (6)$$

In Table II we list the values of the quantum yield calculated from Eq. (4) using the parameters $L = 4.5$ μm and $A = 0.16$ deduced from Eqs. (5) and (6) using the experimental values of $\varkappa_t$. The parameter $\varepsilon = 5$ eV is given by $\varepsilon = 2.8E_g + 0.75$, where E_g is the width of the band gap.[1] The values of the absorption coefficient μ are taken from Ref. 27. Good agreement is observed between the experimental and calculated values of the quantum yield of a $GaP_{0.4}As_{0.6}$ single crystal in the spectral range beyond the K absorption edges of Ga and As. The high value of the calculated quantum yield at $\lambda = 0.07$ nm, compared with the experimental results, demonstrates the need to allow for the energy carried away by the escaping fluorescence in the spectral range up to the absorption edges.

A comparison of the experimentally determined diffusion length L of thermalized electrons, the depth of penetration of x rays $1/\mu$, and the thermalization length L_t shows that in the case of the x-ray photoelectric effect in NEA emitters the thermalization of electrons should be more advanced than in the case of secondary electron emission in the strong absorption region. On the other hand, the thermalization of electrons excited by primary photoelectrons created directly by x rays should occur at all the minima of the conduction band. This demonstrates that additional information on-the energy structure of the conduction band and on the electron scattering processes in the band bending region can be obtained by investigating the characteristics of the x-ray photoeffect and particularly the energy spectra of the emitted electrons.

The angular dependence of the quantum yield $\varkappa_t(\varphi)$ illustrated by the data in Table III is described well by Eq. (4).

The reduction in the quantum yield for a glancing angle of $\varphi = 7°$ when the wavelength is $\lambda = 0.25$ nm can be explained allowing for the fraction of the energy carried away by the primary and "hot" electrons, because the assumption made above of complete thermalization of electrons at low angles is not satisfied. Moreover, at angles $\varphi < 7°$ we have to allow for the reflection and refraction of x rays.

In the range of low angles, where the reflection and refraction effects are manifested, the probability of absorption of x rays is described by the function

$$F(x)dx=[1-R(\varphi)]\sigma(\varphi)\exp(-\sigma(\varphi)x)dx, \qquad (7)$$

where the factor $[1 - R(\varphi)]$ allows for the fraction of the energy absorbed in the medium and the coefficient $\sigma(\varphi)$ determines the extinction length. Then, Eq. (4) for the quantum yield can be written in the form

$$\varkappa_t=An_0(1-R(\varphi))(1+\sigma(\varphi)/L)^{-1}. \qquad (8)$$

The expressions for $R(\varphi)$ and $\sigma(\varphi)$ are known from the theory of dispersion of x rays.[28] A detailed discussion of this topic can be found in Ref. 29 and 30.

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