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Pico-femtosecond image-tube photography in quantum electronics*

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Abstract. **The possibility of experimental achievement of the time resolution of image-converter tubes (ICTs) corresponding to the theoretical limit of 10 fs is considered as applied to quantum electronics problems. A new generation of ICTs with a temporal resolution of 200 – 500 fs has been developed for recording femtosecond laser radiation. The entirely new devices based on time-analysing ICTs such as femtosecond photoelectronic diffractometers, have been created for studying the dynamics of phase transitions in substances using diffraction of electrons with energies ranging from 20 to 40 keV.**

Keywords: *image converter tubes, diffractometer, femtosecond technologies, laser plasma, nonlinear and fibre optics.*

1. Introduction

Ultrafast photoelectronics using time-analysing image converter tubes (ICTs) occupies a special place among traditional methods and measuring techniques (oscilloscopy, autocorrelation methods, videography, and optomechanical photography) employed in quantum electronics. The uniqueness of the recording properties of ICTs lies in their ability to record simultaneously a large amount of spatial information (up to $10^3 - 10^8$ elements with a minimum resolvable size of 20 – 30 μm). In principle, a linear dependence between the number of electrons emitted by the photocathode and the number of photons in the incident electromagnetic radiation is provided at each point of the image (the dynamic range of the external single-photon photoeffect exceeds six orders of magnitude).

The sensitivity of modern photoelectron detectors equipped with image intensifiers with a gain of $10^3 - 10^6$ is sufficient for detecting each individual photoelectron emitted from the photocathode. This means that, for example, in the case of a multialkaline photocathode, a spatially resolvable element will contain only 5 – 10 photons at a wavelength of 500 – 600 nm.

The modern classical (Ag – O – Cs, Na – K – Cs – Sb) and multicomponent (Au + Cs_3Sb, CsI + Cs_3Sb) photocathodes as well as photocathodes with a negative electron affinity (InGaAs/InP) make it possible to study fast processes in the spectral range covering X-rays (0.1 – 1 nm), UV (115 – 350 nm), visible (380 – 780 nm), and near-IR (850 – 1600 nm) emission. The spectral sensitivity of photocathodes can be considerably extended to the red region of the spectrum using multiphoton photoeffect and higher incident intensities.

Apart from the features listed above, an ICT possesses an extremely high time resolution (the time resolution achieved experimentally at present is 180 – 500 fs). Due to the virtually inertia-free external photoeffect, the two-dimensional image of a fast process formed on the ICT photocathode is transformed into its photoelectron replica, thereby converting from the optical range to a higher frequency range characteristic of electrons with energies of several tens of kiloelectronvolts (de Broglie wavelengths are hundredths or thousandths of a nanometer). Basically, this means that the time resolution of a photoelectron information path exceeds that of a purely optical path by several orders of magnitude.

In actual practice, the maximum attainable time resolution of ICTs is limited by the spread in the time of flight of photoelectrons between the photocathode and the phosphor screen, which is directly proportional to the spread in the initial velocities of photoelectrons and inversely proportional to the electric field strength at the photocathode. The broadening of electron wave packets on their path from the photocathode to the phosphor screen due to Coulomb repulsion of electrons and aberration of focusing lenses and deflecting systems considerably affect the time resolution.

Finally, time-swept photoelectron images (with maximum phase velocities exceeding the velocity of light in vacuum) can be stored in a computer, processed, and visually represented.

The Russian scientific school has made a substantial contribution to the development of the physical principles of image-tube recording of fast processes (Zavoiskii and Fanchenko [1]), to the design and development of time-analysing ICTs and image-converter cameras (ICCs) (Butslov, Stepanov, et al. [2]), to their applications in pulsed light techno-logies, nuclear physics, high-current electronics, and plasma physics (Vanyukov [3], Nesterikhin [4], Pergament et al. [5], et al.). Russian scientists were the first to predict the possibility of achieving the 10-fs limit in the time resolution of electrooptical chronography at the IV Interna-

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tional Congress on High-Speed Photography in Köln in 1958 [6]. This formed the basis of meticulous and still ongoing work aimed at achieving this time resolution.

2. Picosecond ICTs of the PIM-UMI series in laser experiments

By the end of the 1960s, domestic ICTs based on the PIM-UMI image tubes series (Fig. 1a) were widely used in quantum electronics and nonlinear optics. These devices provided extremely rich space – time information in the study of discrimination and self-locking of axial modes in ruby and Nd: glass lasers, in photographing the near and far fields of laser radiation, and in recording the spectra, coherence and wave front.

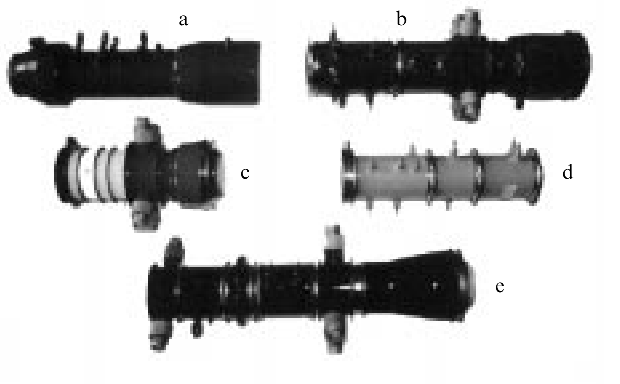


Figure 1. Domestic pico–femtosecond ICTs (image intensifiers are not shown): PIM-3, 1949 (a), PV-001, 1978 (b), BShchV-4, 1989 (c), PIF-01, 1991 (d), PV-FS, 2001 (e).

ICTs were used in a series of studies of laser sparks, stimulated Brillouin scattering (SBS) excited by laser radiation in gaseous nitrogen at a pressure of 150 atm, and observation of the kinetics of self-focusing of laser radiation in nonlinear liquids. These experiments became possible owing to high temporal (smaller than 500 ps) and spatial resolution of the cameras [7] developed by us on the basis of PIM-UMI tubes as well as to the suitability of these cameras for laser experiments (the internal trigger delay was less than 100 ns with an instability of ±0.1 ns).

When axial mode locking was realised in ruby and Nd::glass lasers, the measurement of the space – time structure of ultrashort laser pulses was required. The actual duration of these pulses could be measured neither by oscillographic methods (due to their ultimate time resolution of no better than 50 – 100 ps), nor by the optomechanical photography methods (with a resolution limit of about a few nanoseconds). Autocorrelation techniques, which have, in principle, a femtosecond time resolution, could not solve the problem of measurement of the actual space – time structure of single pulses. The information on the temporal intensity profile for femtosecond laser pulses can be obtained using spectrally-resolved optical gating. These spectral measurements are based on cross correlation, which can be realised either by using the third-order susceptibility (optical Kerr effect) or by generation of harmonics.

By this time, the maximum sweep rate in our electro-optical equipment had been increased to 5×10^9 cm s^{-1}, which provided, in principle, the time resolution equal to 5 ps, while the internal trigger delay was reduced to 20 ns with an instability of ±0.05 ns [8].

Our experiment [9] on the study of the temporal fine structure of the output of a simultaneously Q-switched and self-mode-locked Nd:glass laser (using a bleachable dye) ruled out any ambiguous interpretation of the experimental results available (obtained by using oscilloscopes and autocorrelation). While the oscillograms displayed a regular train of single pulses on the axial mode interval, the ICT recordings showed under the same conditions a rather unstable lasing picture, which varied from shot to shot. It followed from the ICT recordings that each 'single' pulse recorded on the oscillograph contained several subpulses of 10 – 15 ps duration separated by ~ 330 ps.

Since single laser pulses of duration smaller than 10 – 20 ps could not be detected using PIM-UMI tubes, the presence of shorter pulses in laser radiation became disputable. The Zavoiskii – Fanchenko approximate formulas [1, 10] indicated that the limiting resolution of PIM-UMI image tubes determined by chromatic aberrations of the cathode lens should be of the order of a picosecond: the minimum duration of a photoelectron pulse is $\tau_{\mathrm{ch}} \sim 10^{-11}\alpha/E$ (E is the electric field strength at the photocathode in esu and $\alpha = 1 - 5$ is a coefficient which depends on the shape of the distribution of the initial energies of photoelectrons). In PIM-UMI tubes, the electric field strength at the photocathode was 0.3 – 0.6 kV cm^{-1} (~ 1 – 2 CGSE units).

We proposed a mode beat method [11, 12] for measuring the limiting time resolution of ICTs and precise time calibration of sweep rates using images of a narrow slit. The essence of the method lies in ICT recording of sine-modulated (in time) laser radiation with a known period and modulation depth, which allowed a very accurate calibration of the temporal sweep of the ICT. The half-width of the temporal instrumental function can be calculated from the convolution equation for the input signal with the instrumental function.

The mode beat method proved to be indispensable in all the cases when it was difficult to form regular and reliably reproducible optical pulses with duration much shorter than the predicted half-width of the instrumental function. The time resolution of PIM-UMI tubes was measured using a Nd:glass laser generating either two axial modes separated by a controllable spectral interval or single ultrashort pulses [13]. The measurements performed at different accelerating voltages (varying from 10 to 20 kV) across the tubes by illuminating the Ag – O – Cs photocathode by radiation at the fundamental and second harmonic frequencies of a Nd:glass laser showed that the half-width of the temporal instrumental function of an ICT equipped with the PIM-UMI image tubes was 15 – 20 ps at 1060 nm and 30 – 40 ps at 530 nm. Thus, the assumption that the spread in the initial energies of photoelectrons is the main factor limiting the half-width of the temporal instrumental function was confirmed experimentally for the first time.

3. New generation of subpicosecond ICTs with an accelerating grid

By the end of 1970s, a new generation of subpicosecond ICTs specially intended for detecting fast processes in laser experiments had been created in Russia on the basis of PIM-UMI devices [14]. In new tubes (from UMI-93M to

PV-001), the field strength in the vicinity of the photocathode was increased to 30 – 60 kV cm^{-1} (i.e., by a factor of 100 as compared to PIM-3) by using an accelerating grid of a unique structure. Commutation of images was carried out by a compensating gate system and one or two pairs of deflecting plates with coaxial leads having a pass band of up to 3 GHz.

In these tubes, low-resistance Ag – O – Cs photocathodes (surface resistance of the order of 10 Ohm $\square^{-1}$) sensitive in the spectral range from 115 to 1550 nm were used along with two-component ($\text{Au} + \text{SbCs}_3$) photocathodes which are sensitive both in the visible and soft X-ray (0.1 – 1 nm) spectral regions. The output phosphor screen was mounted on a fibre-optic plate, and microchannel plates were used instead of bulky image intensifiers with magnetic focusing.

The electron traps mounted in front of the phosphor screen made it possible to considerably improve the signal-to-noise ratio in the swept images. Instead of supersensitive films used earlier for photographing images from the ICT screen, we successfully tested image readout systems based on ultrasensitive SIT TV cameras and, later used CCD arrays which could detect either photons or electrons.

Dynamic tests of electrooptical cameras and information systems based on a new generation of subpicosecond ICTs were performed on specially designed calibrating laser stands generating sine-modulated and single pico- and subpicosecond pulses [15, 16]. The mode beats of a Nd:glass laser with a minimum period of 1.4 ps and the maximum modulation depth up to 40 % were detected in 1976 using an UMI-93M ICT (the sweep rate $V = 5 \times 10^{10}$ cm s^{-1}, $E =$ 40 kV cm^{-1}) [17]. The estimated time resolution was close to 0.7 ps. The same camera was used for measuring the output pulses from a passively mode-locked Nd:glass laser with a fast-relaxation dye [18]. The minimum duration of a single laser pulse was found to be 1.5 ± 0.5 ps.

To directly measure the instrumental function of a PV-001 image tube (Fig. 1b), we built a quasicontinuous colliding-pulse femtosecond Rhodamine-6G dye laser pumped by a cw argon ion laser [19]. The half-width of the instrumental function upon exposure of an ICT by single 90 – 120-fs pulses at 610 nm was 0.9 ps ($E = 30$ kV cm^{-1}, $V =$ 2×10^{10} cm s^{-1}, Ag–O–Cs photocathode). The instrumental function was narrowed to 0.7 ps when the photocathode was illuminated by pulses of duration less than 500 fs emitted by a $\text{LiF} : F_2^-$ laser at 1.15μm [20].

One of the problems awaiting the solution with the help of picosecond ICTs was to analyse the possibility of their application for studying IR lasers emitting beyond the range of the spectral sensitivity of photocathodes determined by the single-photon photoeffect. We demonstrated the possibility of detecting picosecond pulses emitted by an erbium-doped yttrium-aluminium garnet laser operating in the active mode locking regime at a wavelength of 2.94 μm [21]. The Ag – O – Cs photocathode of the PV-001 image tube reliably detected radiation pulses at the fundamental frequency as well as at frequencies of the second (1.47 μm) and fourth (0.73 μm) harmonics. The threshold sensitivity of detection was 10^8, 10^6, and 10^3 W cm^{-2}, respectively. In these experiments, we estimated the value of nonlinear photoresponse, which was 2.6 ± 0.3 for the second harmonic and 4.6 ± 0.5 for the fundamental frequency of the laser.

Using the new-generation time-analysing ICTs, a variety of experiments were performed and valuable space–time information was obtained. The Er-doped fibre lasers emitting in the IR range (1.55 μm) and intended for the use in ultrafast fibre-optic communication systems were studied in Ref. [22]. The light amplification upon nonlinear interaction of counterpropogating waves in a single-mode fibre was observed in Ref. [23] and the decay of picosecond laser pulses upon self-switching of light in tunnel-coupled optical waveguides was studied in Ref. [24]. The fluorescence decay was detected in reaction centres of photosynthesising organisms [25]. The X-ray radiation emitted by a plasma formed by focusing single 1 – 3-J, 10-ps pulses from a high-power Nd : glass laser on a flat titanium or carbon target placed in a vacuum chamber was photographed in Ref. [26].

4. Femtosecond photoelectron technologies

The advent by the early 1990s [27] of lasers generating pulses of duration of tens or several femtoseconds stimulated new studies in electronics and optoelectronics, in fibre and nonlinear optics, physical chemistry, and synthesis of new materials with pre-programmed properties and molecular dynamics. The importance of the development of direct methods of ICT diagnostics of fast processes with a femtosecond time resolution has become obvious. To solve this problem, the Photoelectronic Department was organised in 1989 at the General Physics Institute, Russian Academy of Sciences on Academician A M Prokhorov's initiative. The work of the department was concentrated on the development of femtosecond photoelectronics, which was a new interdisciplinary trend in scientific research, and included studies in the physics of photoemission, photocathodes of both classical type and that with a negative electron affinity [28], electron optics and mathematical simulation [29], the physics of femtosecond lasers [30], electrovacuum technology and electron-optic instrumentation [31], as well as the computer output, input, and image-processing of time-resolved photoelectronic images.

A new attempt to improve the time resolution of ICTs was made together with Niu [32], when a 50-fs image tube with a cylindrical focusing lens and a compensator of the spread in the tangential components of the initial energies of photoelectrons was simulated on a computer. The experimental model of this ICT constructed at All-Union Research Institute of Optical and Physical Measurements (see Fig. 1c) was used to measure the half-width of the instrumental function, which was found to be 500 ± 50 fs in the single-shot linear sweep regime ($E = 60$ kV cm^{-1}, $V = 1.2$ $\times 10^{10}$ cm s^{-1}) [33]. The discrepancy between the experimental and theoretical results showed that the fabrication technology of ICTs has to be perfected with concerted efforts of all participants of the research team (from mathematicians and experimental physicists to the fabrication engineers and technicians). The necessity for recreating the old technology (which was partially lost) and for developing a new theoretically substantiated and reproducible technology of manufacturing femtosecond image tubes has become obvious.

By the mid-1990s, a compact, flexible, and closed R&D pilot project was set up (from technical assignment to the end product) at the General Physics Institute for developing and manufacturing experimental specimens of femtosecond image converter tubes and cameras. The ICTs of the PIF-1 type (Fig. 1d) and PV-FS (Fig. 1e) manufactured by us at present are globally competitive [34]. They have the following parameters: the size of the input field of the photoca-

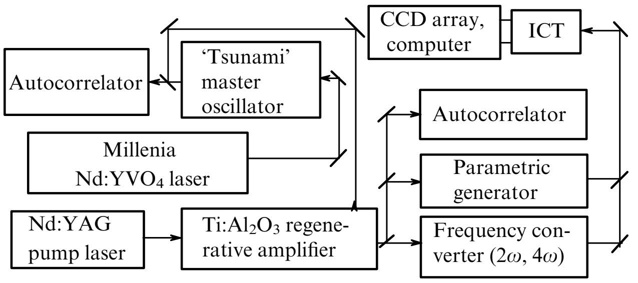


Figure 2. Block diagram of a femtosecond laser setup. The radiation parameters of the master oscillator: pulse duration 60 fs, pulse repetition rate 82 MHz, wavelength 800 nm; output radiation parameters of the amplifier: pulse duration 120 fs, pulse repetition rate 10 Hz, peak energy 1 mJ, covered spectral range 210 – 1600 nm.

thode is up to 4 mm ×20 mm, the spatial resolution relative to the plane of the input photocathode is 30 – 50 pairs of lines per millimetre, the scale of the electrooptical cathode phosphor screen image is 0.7 – 2, the spectral range of sensitivity is 115 – 1600 nm, and the equivalent background noise intensity does not exceed 10^{-7} lm cm^{-2}. Note that a pulsed mode powering of the gap between the photocathode and the grid is provided in the PV-FS ICT. For the electric field strength of 300 kV cm^{-1} achieved at the photocathode of this image tube, the calculated time resolution does not exceed 100 fs.

To determine the temporal characteristics of ICTs and ICCs, a femtosecond laser setup was put in operation at the Department of Photoelectronics in 1995 (Fig. 2). This setup was used to measure the maximum time resolution (400 fs) of the synchroscan camera [35] with the PIF-01 tube. The synchroscan operated by accumulating $10^3 - 10^4$ images of pulses of duration of 60 fs emitted by a Ti:sapphire laser with a pulse repetition rate of 82 MHz.

The possibility of forming femtosecond electron beams in ICTs opened a new field of its application as an electron gun of a photoelectronic diffractometer intended for studying the fundamental properties of matter through the observation of the dynamics of chemical reactions in a substance by the electron diffraction method with a high time resolution [36]. Earlier, Mourou and Williamson [37] used 100-ps, 20-keV electron pulses formed in a time-analysing ICT to observe the electron diffraction pattern from an aluminium film of thickness 150 Å in the transmission mode. We also tried to use a femtosecond electron gun from a time-analysing ICT of the PV-001 type [38] for the diffractometer, but it became evident after several attempts that the image tube functions (generation and measurements of duration of a photoelectron beam) and electron-diffraction functions (formation of an electron beam with the required divergence and obtaining of diffraction patterns) can be combined only in a special device – a femtosecond diffractometric camera on the basis of a femtosecond photoelectron gun (FPG).

Our first FPG was calculated on a computer using the specially worked out software package, designed and manufactured at the Department of Photoelectronics of the General Physics Institute, Russian Academy of Sciences in 1996 [39]. According to calculations, the gun provided the formation of a photoinduced 500-fs electron pulse with an energy up to 30 – 40 keV, and an energy spread less than 0.5 eV. The electron pulse was characterised by an angular divergence less than 0.5° and was focused on the target in a spot of diameter 0.7 mm (by the 1/e level).

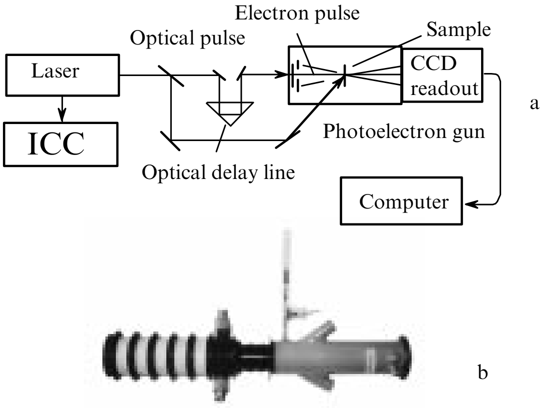


Figure 3. Block diagram of experiments on femtosecond electron diffraction (a) and the appearance of the photoelectron gun (b)

The block diagram of dynamic experiments with FPG is shown in Fig. 3a. We measured the time profile of a photoelectron pulse (Fig. 4) formed in the FPG and showed that a single pulse contains up to 10^3 photoelectrons. To verify the technical parameters of the FPG developed by us, we used an aluminium film of thickness 300 Å. First, we recorded a static diffraction pattern with the help of a standard electron diffractometer. In this case, a 30-keV electron beam was passed through the film for 7 s at a current of 50 μA (Fig. 5a). Then, the same sample was placed in the FPG and the diffraction pattern was recorded (Fig. 5b) by accumulating 4×10^4 photoelectron pulses of duration 500 fs with a repetition rate 82 MHz and containing $\sim 10^3$ electrons per pulse. This means that the sensitivity of the FPG developed by us is almost six orders of magnitude higher than that for a standard electron diffractometer, while its operation speed is higher almost by 13 orders of magnitude!

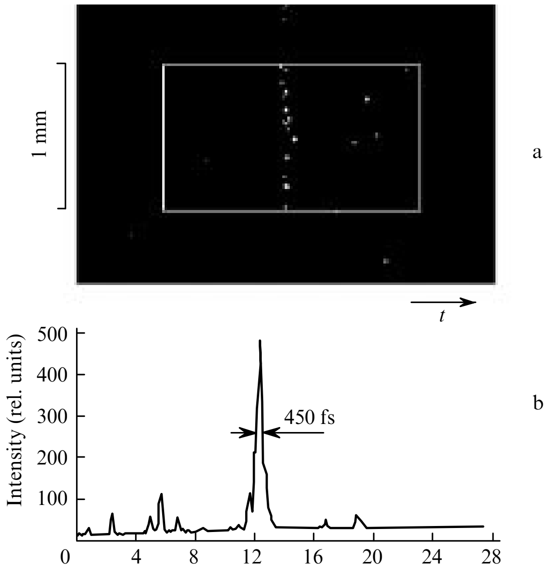
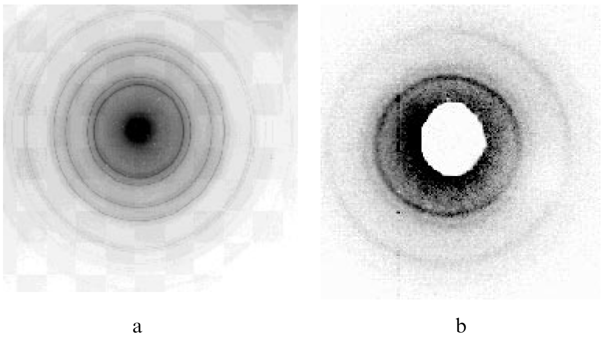


Figure 4. Time sweep of a 500-fs pulse generated by FPG (a) and the corresponding microdensitogram (b).



a b

Figure 5. Diffraction pattern of an aluminium film of thickness 300 Å recorded with a standard electron diffractometer (a) and with a 500-fs photoelectron gun (b).

5. Conclusions

The ultrafast image-tube diagnostics is an efficient experimental tool for solving a number of problems in quantum electronics. At the stage of development of the laser physics, ICTs with a time resolution of 10 ps provided direct recording of fast processes in real time. To preserve the advantages of this method of investigation in modern laser experiments in which the minimum duration of laser pulses has been reduced from picoseconds to femtoseconds, the time resolution of ICTs should be improved by at least an order of magnitude. Many years of experience have proved that this problem cannot be solved using obvious methods (e.g., only by increasing the field strength at the photocathode).

One of the possible methods for improving the time resolution of an ICT by one or two orders of magnitude was proposed by Kuznetsova [40]. In the new method, a special algorithm is used for processing the data obtained for an ultrashort optical pulse during its spectral sweep in an ICT with a high spectral resolution. The procedure of reconstruction of picosecond pulses is considered in detail in Ref. [41], where the potentialities of the method are demonstrated by using computer simulation.

In his lessons on femtosecond photography, Fanchenko [10] proposed that the experiments on the creation a 10-fs ICT should be carried out on the Moon! First, a high vacuum is available there. Second, solar annealing in a high vacuum and the absence of dust guarantee the absolute cleanliness of all the surfaces of the instrument and the solution of many technological problems.

However, returning to the solution of terrestrial problems, we should emphasise that the main problem existing at this stage is associated with analysis and inclusion of more complicated and sparsely studied factors determining the time resolution of ICTs. This primarily refers to the physical properties of photocathodes and photoemission in the regime of strong electric fields acting on the photocathode.

The energy levels of the photoemitter will be strongly distorted as a result of the penetration of an external field into thin photocathode films (having a thickness of hundreds angstroms). The anomalous dependence of the time resolution on the electric field strength near the photocathode was noted by us earlier (for example, in measurements of the instrumental function of the PV-001 tube with the help of 300-fs single pulses emitted by a Rhodamine-6G dye laser at 615 nm) [42].

In tubes with an accelerating grid, the work function of the photocathode becomes much smaller, and the spectral sensitivity is shifted to the red (Schottky effect). In addition, the linear field distribution may be distorted in the gap between the photocathode and the grid due to the effects caused by spatial charge and the potential induced by a finite resistance of the photocathode. It is high time to study meticulously these and many other factors determining the actually attainable limit of time resolution of ICTs.

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