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Two-electron pulses of a photomultiplier and two-photon photoeffect

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***Abstract*. Pulses arising when the cathode of a photomultiplier emits two electrons are studied. It is shown experimentally that under certain conditions, emission of individual photoelectrons forming a pair cannot be considered as independent but should be described as a single two-photon photoeffect.**

Keywords: *photomultiplier, two-electron pulses, two-photon photoeffect.*

1. Introduction

The term two-photon photoeffect is usually applied to a process in which absorption of two photons leads to the transition of a single electron from a bound state into a state of continuous spectrum. In 1964, Artem'ev [1] reported the experimental observation of another type of two-photon photoeffect, in which absorption of two photons is accompanied by the transition of two electrons into a state of the continuous spectrum. The statement that the observed effect is indeed a two-photon photoeffect rather than a simple combination of two one-quantum processes is confirmed by a very high probability of two-electron emission by the photocathode, exceeding by several orders of magnitude the squared probability of single-photon emission within the same (very short) time interval. In Ref. [1], this high probability was attributed to the coherence of light incident on the photocathode.

Later and independently of Artem'ev's work, a similar two-photon photoeffect (emission of two photoelectrons caused by the simultaneous absorption of two photons) was reported in the work [2] by Klyshko and Penin, where the photocathode of a photomultiplier tube (PMT) was illuminated by a light field in an essentially quantum (nonclassical) biphoton state. However, no anomalously high probability of such photoeffect was discovered. Moreover, the quantum efficiency of detecting two photons was assumed by the authors of Ref. [2] equal to the square of the quantum efficiency of single-photon photoeffect. This assumption formed the basis for the single-channel method of reference-free calibration of the PMT quantum efficiency. The conclusion that the observed phenomenon should be called the two-photon photoeffect (rather than the combination of two single-photon photoeffects) was based in [2] on the special two-photon statistics of light hitting the PMT cathode. An important difference between papers [1] and [2] was that they used different photocathodes. In Artem'ev's paper, a cesium – antimonide photocathode was studied, while Penin and Klyshko worked with multialkali photocathodes. The results by Artem'ev were reproduced in Ref. [3] for the same cesium – antimonide photocathode. The high probability of the effect was confirmed; however, it was shown to be not related to the coherence of light. The origin of the observed phenomenon remained, therefore, unexplained.

Summarising, we can conclude that Refs [1] and [2] were the first experimental observations of two-photon two-electron photoeffect; however, it remained unclear whether the quantum efficiency of detecting two photons within a short time interval should be always equal to the square of the quantum efficiency of detecting a single photon. If the answer is 'no', then, under what conditions can one detect photon pairs with a considerably higher probability? What is the relation between the probability of such a two-photon photoeffect and the statistics of light illuminating the photocathode? All these questions can be answered experimentally by studying two-electron pulses of a PMT, which is the subject of the present work.

2. Classical and nonclassical two-electron pulses of a PMT

A PMT is a device that enables quantum measurement over an electromagnetic field. For a better clarity of further consideration, it is useful to consider the process of detecting photons in a PMT in the general terms of the quantum measurement theory. Semitransparent photocathodes usually have a very small thickness (about 10 nm), which is comparable to the free path of an electron in the photocathode material. The problem of the electron exit from the photocathode should be therefore considered as the problem of the electron passing through the potential barrier at the boundary between the photocathode and vacuum. From the viewpoint of the quantum measurement theory, a measurement is considered as an interaction between the quantum system under measurement and a classical device. The very exit of the electron from the

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cathode, apparently, is not yet a measurement. It is only the interaction of the electron with the first dynode that is actually a quantum-mechanical measurement. Hence, it follows that two-electron PMT pulses can be of principally different nature. If each electron of a pair was detected independently of the detection of another electron, we deal with a simple overlap of two single-electron pulses, which cannot be detected separately only due to a finite resolution time of the detection system. The probability of such a two-electron pulse appearing within a sufficiently short time interval should be apparently equal to the squared probability of a single-electron pulse detection. Below, for the sake of brevity such two-electron pulses will be called classical ones. If the second photoelectron arises before the first one interacts with a classical measurement device (the first dynode), the problem should be treated quantum-mechanically as a two-electron one. The probability of a two-electron pulse is no more necessarily equal to the squared probability of a single-electron pulse, and this statement will be further proved in experiment. Let us call such two-electron pulses nonclassical ones.

Consider first classical two-electron pulses, which can be also of interest for the experimental study of photon statistics. The counting rate of such pulses considerably depends on the time resolution of the setup. This is clear from Fig. 1a, which shows pulses of various shapes from a FEU-64 PMT, recorded with a Tektronix TDS 3052 oscilloscope (frequency bandwidth 500 MHz). Closely spaced single-electron pulses, which are well resolved by the oscilloscope (the curve $2 \times$ 1e1ph), may be interpreted by the subsequent electronics as a single two-electron pulse if, for instance, the pulse amplitudes are measured with a multichannel analyser (MCA). This is because the time resolution of the system is often partly sacrificed in order to increase the accuracy of the amplitude analysis.

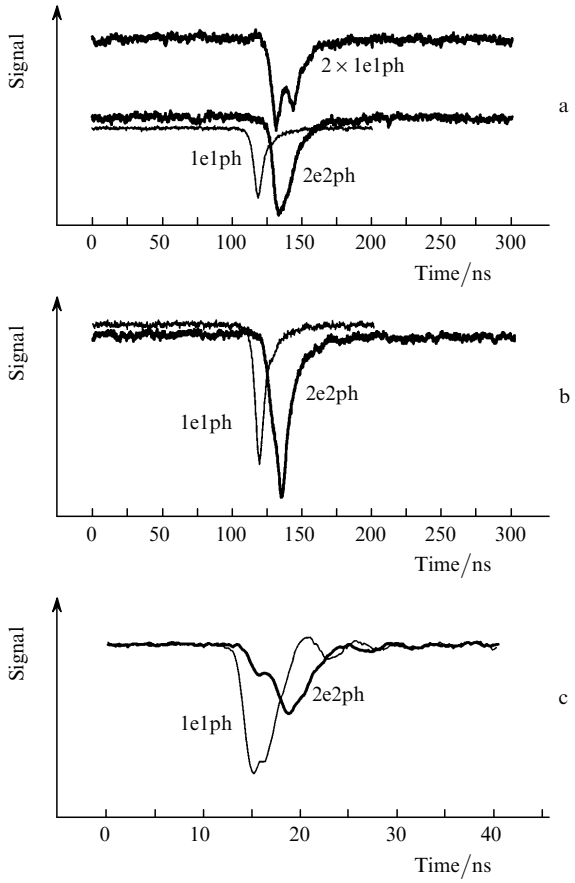

Figure 1. Single-electron (1e1ph), two overlapping single-electron ($2 \times$ 1e1ph), and two-electron (2e2ph) pulses of a FEU-64 PMT (a), as well as a single-electron pulse and a two-electron pulse with the stretched leading edge from FEU-64 PMT (b) and PhilipsXPH277 PMT (c).

One can see from Fig. 1a that there also exist strongly overlapping single-electron pulses separated by time intervals that are much less than the single-electron PMT pulse duration (curve 2e2ph). In this case, the above-mentioned time separation has a strong effect on the shape of the two-electron pulse. In particular, when the time interval between single-electron pulses is close to the rise time of a single-electron pulse, pulses with stretched leading edges appear, shown in Fig. 1b. Such two-electron pulses already cannot be resolved even with an oscilloscope with the highest resolution; however, it does not mean that such pulses are nonclassical.

Indeed, for a two-electron pulse to be classical, the time interval between the escapes of single electrons from the photocathode should exceed the electron time of flight τ_0 from the photocathode to the first dynode. This time can be easily estimated since the voltage U between the cathode and the first dynode is known, and the initial velocity of the photoelectron is negligibly small,

$$\tau_0 = l\left(\frac{2m}{eU}\right)^{1/2}. \tag{1}$$

Here, l is the distance from the cathode to the first dynode; e and m are the electron charge and mass. For dynode PMTs, estimate (1) yields a time of $\sim$1 ns, which is always less than the duration of a PMT single-electron pulse but can be comparable with the pulse rise time. This is the case for the PhilipsXPH277 PMT, whose two-electron pulses with stretched leading edges are shown in Fig. 1c. By assuming that $l = 10$ mm and $U = 180$ V (the PMT has 14 dynodes, a uniform voltage divider, and operates under a bias voltage of 2700 V), we obtain $\tau_0 = 2.5$ ns. This means that nonclassical two-electron pulses should be searched either among pulses with the same shapes as single-electron ones but with approximately doubled amplitudes, or among two-electron pulses with stretched leading edges, provided that the rise time of a single-electron pulse is less than or close to the electron time of flight between the photocathode and the first dynode. All other two-electron pulses, similar to the ones shown in Fig. 1a, are classical.

3. Critical analysis of papers [1] and [2]

Consider in detail the results of Refs [1] and [2] and the questions that still remain unanswered. It should be noted that both works are thorough and well-designed, and their experimental results should be trusted. Nevertheless, these results do not provide a clear and unambiguous picture. This means that we should reconsider the existing results in order to outline the ways for constructing a noncontradictory model of a two-photon photoeffect.

While in Ref. [2] the equality between the two-photon efficiency and the square of the single-quantum efficiency was taken as a starting postulate, in Ref. [1] a considerable increase of the two-photon efficiency compared to the square of the single-quantum one was explained by the

coherence of light. This explanation was based on the fact that the effect was observed only for the photocathode illuminated with spectrally narrowband light and was absent in the case of white-light illumination. This experimental fact is very important for the understanding of the two-photon photoeffect, but its explanation in terms of the light coherence contradicts the observation conditions. Indeed, no special measures were taken in Ref. [1] to provide the spatial coherence of light illuminating the photocathode or to select its polarisation. In other words, the photocathode was illuminated by light with a small coherence radius. Under these conditions, correlation of photons is known to be absent. Even if we assume that the conditions for coherent illumination of the photocathode were satisfied accidentally, due to a very narrow slit of the monochromator and its considerably different transmission for two orthogonal polarisations, it remains unclear how photon correlations could be manifested for sources used in Ref. [1]. As narrow-band sources, the authors of Ref. [1] used PRK-2 and DRSh-250 mercury lamps. The spectral lines of such lamps are broadened because of the gas pressure, so that their width exceeds considerably the Doppler broadening, and the corresponding coherence time is of the order of several picoseconds. This means that the results of the photocurrent-pulse correlation function direct measurement, performed in Ref. [1], should be explained without considering photon statistics. On the other hand, these results indicate that electrons forming a two-electron pulse escapes from the photocathode not simultaneously but with a typical time separation of ~ 1 ns, so that in the case of a PhilipsXPH277 PMT they may well form a pulse with a stretched leading edge.

As we have already mentioned, the fact that the two-photon efficiency is equal to the squared one-quantum efficiency was accepted in Ref. [2] as the starting postulate; however, this postulate is not evident. There are two possibilities here. The first one is that the equality holds true namely for the case of biphoton light and the second one, that it is only true for the particular photocathode type used. Theoretical consideration of the usual two-photon photoeffect (emission of a single electron caused by the absorption of two photons) in biphoton light has been carried out only recently [4] and has shown that the probabilities of this process are considerably different for the cases of thermal and biphoton light. It seems reasonable to assume that the two-photon two-electron photoeffect is of general nature, i.e., can be observed for all types of photocathodes, but its efficiency may depend on the photocathode type. In this connection, it would be interesting to detect the two-photon two-electron photoeffect in a multialkali photocathode for light from a thermal source and to compare it with the similar effect for biphoton light.

4. Experimental

To reproduce the results of works [1] and [2], we used the setups shown schematically in Fig. 2. The influence of the coherence of light on the two-photon photoeffect was tested by using thermal light sources with different coherence times. Both photomultipliers used by us were placed into cooled housings. During the measurements, the photocathode temperatures were $-15 \ldots -25\,^{\circ}\mathrm{C}$, which provided a low level of dark noise (of the order of several counts per second). To separate a narrow wavelength range and to provide spatial coherence of light, we used a three-prism monochromator, whose entrance slit was an aperture of 2 mm in diameter placed at a distance of 380 cm from the first prism (Fig. 2a). Light from the source S was collimated using a second aperture of the same diameter placed directly in front of the first prism. To eliminate stray light, tubes of diameter 18 mm with black inner surfaces were placed between these two apertures, and additional apertures of 5 mm in diameter were inserted between them. This construction caused too much parasite light

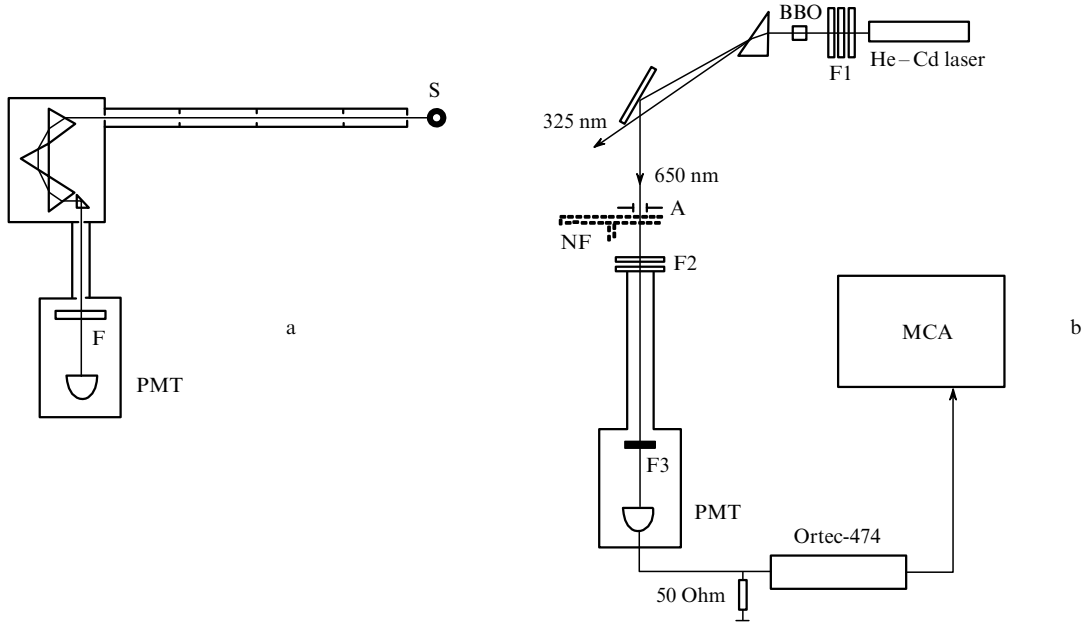

Figure 2. Optical schemes of experiments with illuminating photocathodes by coherent radiation (a) and biphoton light (b): (S) light source; (F) KS-10 filter; (F1) UFS-1 filters; (F2) ZhS-4, KS-10 filters; (F3) ZhS-4 filter; (NF) neutral filter with variable optical density; (A) aperture of diameter 3 mm; (Ortec-474) amplifier; (MCA) Trump-PCI-8K multichannel analyser.

hitting the prisms because of light passing through the first aperture and reflected from the tube sides; in order to avoid it, the tube inner surfaces were covered with black velvet. After passing through three prisms, light was directed by another prism, mounted on a rotation stage, towards another system, light-insulated in a similar way, consisting of two apertures with diameters 2 mm, separated by a distance of 61 cm. The prisms, made of TF-5 heavy flint glass, were equilateral ones. They were oriented in such a way that refraction occurred at nearly Brewster angles. The instrumental function of the monochromator at the wavelength of 650 nm had a FWHM of 5 nm. The monochromator transmission at this wavelength was about 10 %, the degree of polarisation being close to 70 %. Such a construction enabled one to reliably select a desired line out of the spectrum of a gas-discharge lamp and simultaneously provided a diffraction-limited output light beam with the specified polarisation, which was important for studying the influence of light statistics on the observed effects.

For experiments with biphotons, we used the setup shown in Fig. 2b. The amplitude distribution of the output PMT pulses was studied by using an MCA. Biphotons were generated at a wavelength of 650 nm via spontaneous parametric down-conversion of light from a 325-nm pumped He–Cd laser in a BBO crystal with type I phase matching. Emission of a gas-discharge plasma, which caused undesirable background illumination of the photomultiplier cathode, was suppressed by means of three UFS-1 filters of thickness $d = 3$ mm each, placed in sequence before the crystal. Biphoton light was separated from the pump radiation using a prism, an aperture A of diameter 3 mm, and spectral filters F2 (ZhS-4, KS-10, $d = 3$ mm) and F3 (ZhS-4, $d = 2$ mm). A neutral-density filter NF with variable optical density was used to balance the intensities of light while comparing PMT pulse distributions for the cases of biphoton light and the light emitted by the laser tube plasma. In the latter case, the BBO crystal and one of the UFS-1 filters were removed, which provided sufficient illumination of the PMT photocathode by the radiation of the laser tube plasma. The PMT output pulses were inverted, amplified by an Ortec-474 amplifier, and fed to a Trump-PCI-8K multichannel analyser card in a PC.

We used the following simple and efficient way of counting pulses with the stretched leading edge. Such pulses arise especially often in PMTs with large photocathode areas; they reduce the time resolution and spoil the linearity of the PMT response as a function of the incident light intensity because their counting rate has a quadratic dependence on the light intensity. Therefore, some discriminators have a special SRT (slow rise-time reject) regime, in which every PMT pulse is tested for the leading edge slope, and all pulses with the stretched leading edge are not detected. A simple change in the discriminator operation logic provides, on the contrary, the detection of only pulses with the stretched leading edge and to ignore usual single-electron PMT pulses.

Simultaneously with counting pulses with the stretched leading edge, pairs of single-electron pulses were also counted during a fixed short time interval (Fig. 3). The PMT output pulses were split using a matched divider and fed to discriminators A and B, one of which detected only pulses with the stretched leading edge, while the other operated in the usual regime. The discriminator B had a

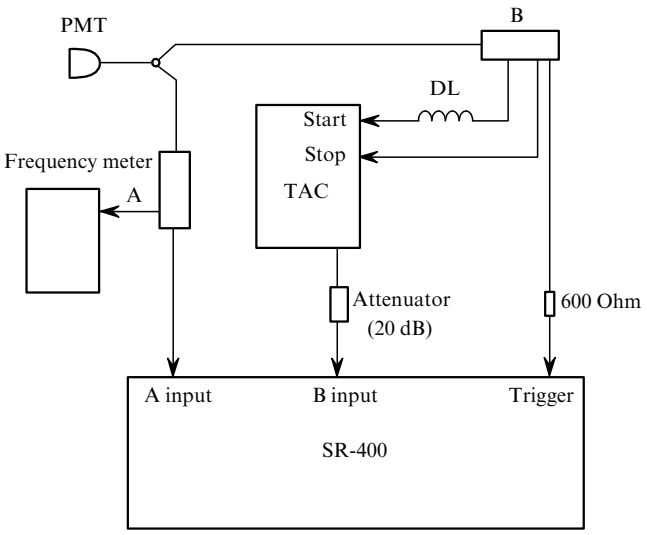

Figure 3. Scheme of the experimental setup for counting two-electron pulses with the stretched leading edges: (A, B) CF-discriminators; (DL) delay line; (SR-400) photon counter.

threshold chosen in such a way that it reliably detected single-electron pulses, which, under all conditions, constituted the majority of all pulses. Discriminator B was also triggered by a sufficiently intense two-electron pulse at its input, but the number of such pulses was always much smaller than that of single-electron pulses. Discriminator A could be switched from the regime of counting pulses with the stretched leading edge to the usual operation regime. Its threshold was chosen in such a way that in the usual regime, the counting rate coincided with the counting rate of discriminator B.

When the input pulse of Ortec-584 discriminators, used in our experiments exceeded a threshold, the discriminators generated three pulses at their outputs; two of them were negative NIM pulses and the third one, a positive TTL pulse. The output signals of discriminator A were fed to a meter of the pulse counting rate, which allowed one to control the illumination of the PMT photocathode, and to the A input of an SR-400 photon counter (Stanford). One of the NIM signals from discriminator B was fed through a delay line to the Start input of a time-to-amplitude converter (TAC). The same signal but without the delay was fed to the stop input of the TAC. A TAC operates in such a way that it ignores a 'stop' signal appearing before the 'start', as well as any repeated 'start' pulse appearing before a 'stop' pulse. For this reason, in our setup the first photon started TAC and the second one stopped it, i.e., the TAC counted all photon pairs appearing within a given time interval. A pulse from the Valid Conversion output of the TAC indicated the detection of a photon pair and was fed to the B input of the SR-400 counter. TTL pulses from discriminator B went to the trigger input of SR-400 and served for measuring single-photon counting rate.

The pulses were accumulated by the counter under the control of a personal computer. The measurement procedure was as follows. After fixing an appropriate level of the PMT illumination, discriminator A was switched into the mode of counting pulses with the stretched leading edge, and their counting rate was measured at the A input of the counter. The counting rate of single-electron pulses was

measured at the trigger input of SR-400, and the photon pair counting rate was measured at the B input. Thus, we could simultaneously measure the counting rates of single-photon events, pairs of single-photon events, and two-photon events. This registration scheme allowed one to check, in a most reliable way, whether two-photon events can have probabilities much exceeding the squared probability of one-photon events. Indeed, let the mean counting rate of single-electron pulses be $r = \eta\bar{I}$, where η characterises the photodetector efficiency and $\bar{I}$ is the mean intensity of light incident on the photocathode. Then the mean counting rate of pairs of single-electron pulses during a time T is (see the Appendix)

$$R_2(T) = \eta^2\bar{I}^2T = r^2T. \quad (2)$$

This formula should also give the counting rate of two-electron pulses with the stretched leading edge if they are classical. Taking the coherence of light into account leads to an additional factor of the order of a unity appearing in Eqn (2) (see the Appendix). Clearly, the integration time for the case of detecting two-electron pulses with the stretched leading edge should not be much different from the duration of a single-electron PMT pulse; hence, Artem'ev's statement [1] can be experimentally verified for two-electron pulses. If the factor standing by the quadratic term in the experimentally measured dependence of R_2 on r for pulses with the stretched leading edge exceeds considerably T (by more than an order of magnitude) upon illumination by narrow-band light and remains of the order of T upon white-light illumination, we will have to conclude that the efficiency of detecting a photon pair is in the first case much higher than η^2, i.e., that two-electron pulses with the stretched leading adge are nonclassical ones. In this case, the photon-pair counting rate at input B should always correspond to formula (2).

Let us now make several remarks on the methods of measurements with white light and on the measurement of the spectral dependence of the output pulse amplitude distribution. In experiments with white light, the PMT photocathode was illuminated directly by a SI6-100 incandescent lamp, whose radiation was attenuated by neutral filters. We tried different versions of illuminating the cathode, based on either focusing the light by means of an objective lens or simply restricting the beam aperture by a diaphragm placed in front of the PMT. In general, the pulse amplitude distribution depends on the size of the illuminated cathode area and the position of the light spot on the photocathode. Most probably, this is caused by slight variations in the amplification coefficient and the electron collection coefficient and requires a certain caution in the interpretation of results. Here, it was very helpful that simultaneously with detecting two-electron pulses, we detected pairs of single-electron pulses according to the method described above. All changes in the amplification coefficient and photoelectron collection coefficient should be reflected in the same way in the number of detected pairs of single-electron pulses and the number of classical two-electron pulses; therefore, a considerable change in the number of the latter with a constant number of pairs of single-electron events could indicate the nonclassical origin of two-electron pulses.

The spectral dependence of the amplitude distribution of output pulses was measured by transmitting radiation of the SI6-100 incandescence lamp through a DMR-4 double prism monochromator and focusing into a 5-mm spot on the PMT photocathode. The instrumental function of the monochromator had a width of 3 nm at 650 nm, $\sim$5 nm at 900 nm and $\sim$10 nm at 1000 nm. We did not take any special measures for providing the spatial coherence and selecting polarisation. The lamp filament temperature measured with an optical pyrometer was 2373 K in all the measurements, both spectral and with white light.

Let us finally describe light sources used in our experiments. As a thermal light source with a sufficiently large coherence time, we used the plasma tube of an ILGN-101 He–Ne laser (the output cavity mirror of the laser was removed). It is known that the gas pressure in such a tube is chosen by the laser manufacturers so that the Doppler broadening dominates and determines the width of the laser amplification line. For a He–Ne laser under normal conditions, the Doppler width is $\sim$1500 MHz, which corresponds to a coherence time of 0.67 ns, if it is determined from the uncertainty relation, and of 0.44 ns, if the Mandel definition of the coherence time is used [5]. Such estimates, although looking trustworthy, are still indirect ones and cannot ensure that the particular laser tube we used is indeed a light source with the given coherence time. The coherence time was also estimated in the course of calibrating a Brown–Twiss interferometer, which was used for other measurements. We obtained a value of the order of magnitude of the above estimates. With the time resolution of the Brown–Twiss interferometer of $\sim$1 ns, the number of photocount coincidences at zero delay exceeded the corresponding number on the edge of the scanned delay range (25 ns) by approximately 10 %. In some experiments, the photocathode was illuminated by attenuated laser light at 632.8 nm. This was achieved by adding the output mirror of the ILGN-101 laser cavity and several neutral filters. As a quasi-monochromatic light source with small coherence time, we used the radiation of the SI6-100 incandescent lamp transmitted through a three-prism monochromator. As a result, we obtained the 650-nm line of width $\sim$5 nm, which corresponds to a coherence time of $\sim$0.045 ps. To provide stationary conditions of the experiment, the setup, including electronics and the light sources, was kept working for at least one hour before the start of the measurement.

5. Single-channel scheme of intensity correlation measurement

The above-described method of detecting two-electron pulses is of certain interest from the practical viewpoint because it allows one to improve the single-detector measurement of photon statistics proposed in 1966 by Morgan and Mandel [6]. This method was based on measuring the counting rate of single-electron pulse pairs with a fixed delay time between the pulses. The pair counting rate is in this case expressed through the integral of the intensity correlation function, in fact, the same way as in (A8). This method has a certain advantage compared to the commonly used Brown–Twiss interferometer. First of all, it has a larger rate of pair counting, since the initial beam is not split in two, and hence, the intensity of light incident on the photocathode is twice as high and the pair counting rate is four times as high. If one also takes into account that splitting of the beam is always accompanied

by light losses at the beamsplitter and that in practice, it is not always easy to balance the counting rates of two PMTs due to the beamsplitter imperfections, slightly different photocathode sensitivities and different illumination, then the benefit in the pair counting rate becomes even more evident. Additional advantages of the single-channel scheme are its simplicity and the absence of complicated alignment.

The main disadvantage of this scheme, which prevents it from being commonly used, is a natural restriction on the measured time intervals between the pulses: these intervals should exceed the duration of the PMT single-electron pulse. While in the Brown – Twiss method one can measure the integral of the whole correlation function, including the range of small delays, where it is most different from the unity, in the method of Morgan and Mandel the integral is taken only over the 'tail' of the correlation function. Therefore, reduction of the time τ determining the lower integration limit is of principal importance. While detecting pulses with the stretched leading edge, we are, in fact, counting pairs of single-electron pulses separated by time intervals equal to the duration of the pulse leading edge, which broadens the possibilities of the single-channel method. Because in this case we speak of the 'classical' overlap of single-electron pulses, a similar effect (arising of pulses with the stretched leading edge) should be observed in the response of a PMT with microchannel plates, whose single-electron pulses are of subnanosecond duration for the rise times being ~ 200 ps. The use of such PMTs considerably improves the time resolution of the single-channel method. As a result, it will enable one to measure coherence times starting from 400 ps. Note that recently methods of quantum optics are actively used in solid-state physics for the study of single quantum dots with typical transition times $\sim 0.7 - 4$ ns [7 – 9] and the use of the single-channel method for such studies is very promising.

6. Results of the study of a multialkali photocathode

Pulse amplitude distributions obtained for a PhilipsXPH277 PMT illuminated by biphoton light and light from a thermal source (gas plasma in a laser tube) are shown in Fig. 4. Note that the maximum, corresponding to the detection of single-electron pulses is shifted towards large amplitudes upon illumination by biphoton light. The counting rates in the two cases were very carefully balanced by means of a neutral filter with variable optical density; therefore, the difference in the two distributions can be attributed to the difference in the statistics of light incident on the photocathode. It is known that the correlation function of photons forming a biphoton has a typical correlation time of ~ 100 fs, i.e., photons arrive at the photocathode practically simultaneously. The arising two-electron pulses should not considerably differ in shape from single-electron ones; however, they should have, on average, the doubled amplitude. This leads to an increase in the probability of observing pulses with large amplitudes and to the corresponding shift of the single-electron peak.

From the curves shown in Fig. 4, the photocathode quantum efficiency can be estimated by the method described in Ref. [2]. Estimates give a value of $\sim 20\,\%$, which noticeably exceeds the passport value of 7 %, especially if one takes into account that the PMT is more than 15 years old. In our experiments, we did not aim at the accurate calibration of the PMT; therefore, it would be wrong to interpret the obtained result as observation of the nonclassical mechanism of photon-pair detection. For accurate calibration, one should use the differential method, similar to the one described in Ref. [2], which would allow one to minimise the systematic errors caused by time instability and small differences in the photocathode illumination by different light sources. However, it is important for us that for the multialkali photocathode, we do not see the anomalous number of two-electron pulses observed in [1], even under illumination with biphoton light containing an extraordinarily large number of correlated photons.

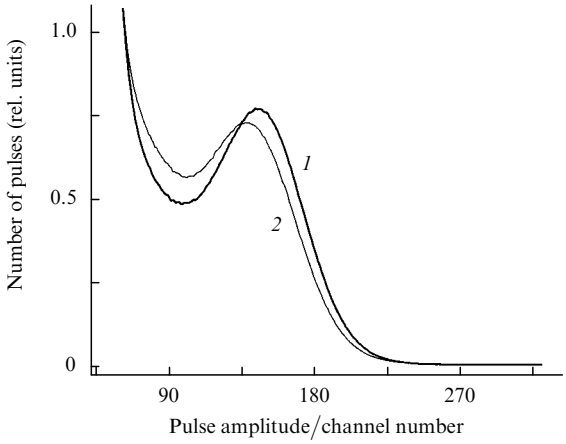

Figure 4. Count characteristics of a PhilipsXPH277 PMT illuminated by the biphoton light (*1*) and the radiation of the plasma discharge of a He – Ne laser tube (*2*). The integral counting rate is 4700 pulses s^{-1}.

Under the same conditions as the ones used for obtaining the curves in Fig. 4, we measured the counting rates of two-electron pulses with the stretched leading edge whose shape in the case of the PhilipsXPH277 PMT is shown in Fig. 1c. No differences between the counting rates in the cases of biphoton light and light from a thermal source have been observed. The counting rates for pulses with the stretched leading edge were also measured by illuminating the PMT photocathode by radiation of thermal sources with various coherence times, as well as by laser radiation (6328 Å He – Ne laser), using the setup shown in Fig. 2a.

The dependence of the counting rate for such pulses on the total counting rate of all pulses (both single- and two-electron) for the PhilipsXPH277 PMT illuminated by white light (incandescent lamp) is shown in Fig. 5a. One can see that this dependence is described with a good accuracy by a quadratic function with the coefficient 7.2 ns, while the duration of a single-electron pulse is 4 ns. The counting rate of pairs of single-electron pulses, which was measured simultaneously with the counting rate of two-electron pulses, is shown in Fig. 5b. Comparing the results for two different light sources, we see that the corresponding differences in the counting rates of two-electron pulses are within the ranges of experimental errors. This result does not look unexpected if one takes into account that the Doppler width of the narrowest spectral line among the lines of thermal sources at our disposal was 1500 MHz, while in the experiments by Morgan and Mandel it did not exceed 200 MHz.

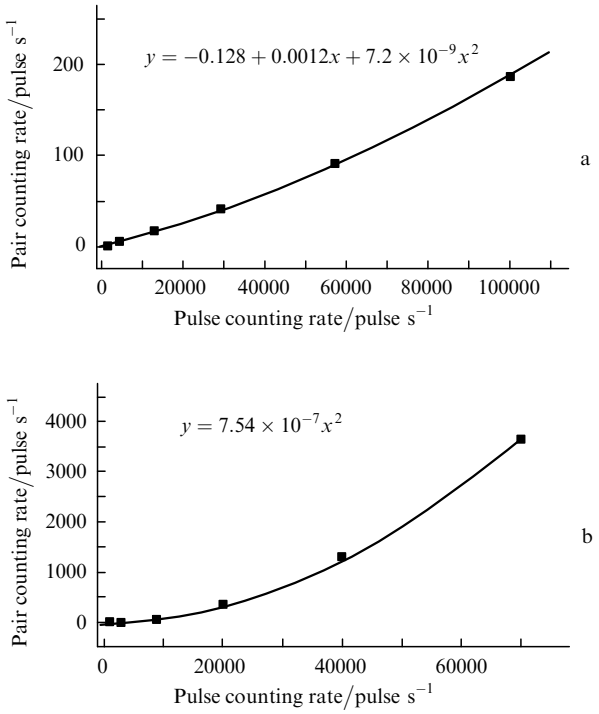

Figure 5. The counting rate of two-electron pulses with the stretched leading edge (a) and the counting rate of single-electron pulse pairs (b) for PhilipsXPH277 PMT as functions of the integral counting rate of all pulses (single- and two-electron) upon illumination by white light. Points are experimental data, curves are theoretical approximation.

Before making any conclusions from the obtained data, it is useful to discuss some measurement details that affect the accuracy of the results. The abscissas in Fig. 5 are the counting rate of all pulses; however, as one can see from Fig. 5a, the counting rate of two-electron pulses is always much lower than the counting rate of single-electron ones; therefore, it is not a big mistake to assume that Fig. 5a shows the dependence of pair counting rate on the counting rate of single-electron pulses, i.e., on the intensity of light incident on the photocathode. A certain measurement error is caused by the choice of the discriminator thresholds, although it seems, at first sight, that this choice is not essential. The formulas always include the counting rate of single-electron pulses, i.e., the product of the detector efficiency (depending, in particular, on the discriminator thresholds) and the intensity of light. Therefore, one can achieve the same counting rate at different discrimination thresholds by adjusting the intensity of light.

This reasoning is indeed valid when the pulse pair counting rate is measured using a TAC; however, when one counts the rate of pulses with the stretched leading edge, the situation is somewhat different. In a measurement with TAC, a set of pairs is selected from the set of detected single-electron pulses, each of which has its amplitude exceeding the discriminator threshold. A pulse with the stretched leading edge is formed by two partly overlapping electron avalanches, so that its amplitude always exceeds the amplitudes of pulses that could be formed by these avalanches separately. As a result, a part of such two-electron pulses could be formed by pulses with small amplitudes, which do not contribute into the single-electron pulse counting rate. The measurement accuracy for the counting rate of two-electron pulses with the stretched leading edge can be improved by introducing a correction to the single-electron pulse counting rate, taking into account pulses with small amplitudes, i.e., plotting the counting rate of such pulses as a function of the total counting rate of single-electron pulses with amplitudes either larger or smaller than the discriminator threshold. For this purpose, we measured the pulse counting rate using the discriminator and the MCA, under exactly the same experimental conditions. The integral pulse counting rate measured by the MCA was always higher than the counting rate measured by the discriminator, since the MCA was counting both single-electron and noise pulses. This means that in the distribution obtained with the MCA, one could mark the amplitude corresponding to the discriminator threshold and estimate the fraction of pulses lost by the discriminator. We obtained corrections of the order of 30 %. While introducing these corrections, it was important to take into account the MCA dead time, i.e., to chose its mode of operation in which the dead time was compensated automatically. It is enough to say that for the integral counting rate of about $\sim 10^5$ pulse s^{-1}, the dead time was approximately 80% of the real time. Points in Figs 5a, b were obtained simultaneously, in a single experiment and for the same level of the photocathode illumination; however, the counting rates plotted along the abscissas differ because an appropriate correction is introduced for two-electron pulses.

Fitting of the experimental data in Figs 5a and b is also different. In the case of pulse pairs (Fig. 5b) we should fit the data by a function that is only quadratic in the pulse counting rate, while for two-electron pulses (Fig. 5a), only fitting with a second-order polynomial has a physical meaning. The linear term in this polynomial accounts for the errors of the discriminator, which sometimes takes for a two-electron pulse an oscillating noise on the tail of a very high single-electron pulse. This term depends on the settings of the CF-discriminator and is considerably reduced if the CF-delay is chosen correctly. Finally, note that the integration time (750 ns) obtained from the quadratic fit for pairs of single-electron pulses turns out to be considerably smaller than the used TAC time sweeping range (2000 ns). Probably, this discrepancy is caused by the effect of the TAC dead time, which reduces the efficiency of pair counting.

Taking all this into account, one can conclude that the detection probability of two-electron pulses observed by us does not differ from the probability of two single-electron pulses overlapping, since the accuracy of our measurements does not allow one to attribute the difference between the expected integration time ($T = 2 - 4$ ns) and the observed integration time (7 ns) to the nonclassical nature of two-electron pulses. This, certainly, does not mean that there are no nonclassical pulses in this experiment; moreover, one can say that there should always exist pulses formed by two electrons exiting the photocathode within the time interval smaller than their time of flight to the first dynode. The obtained result only means that the escape probability for two electrons is not anomalously high.

There exists another independent way of searching for nonclassical two-electron pulses of a photomultiplier, based on studying the dependence of the PMT pulse amplitude distribution on the wavelength of the illuminating quasi-monochromatic source. The idea of this method is rather simple. If nonclassical two-electron pulses have indeed an

anomalously high probability, this should be explained in terms of some other photoeffect mechanism, which, most probably, would have the wavelength dependence different from the one-quantum one. This means that the output pulse amplitude distribution should depend on the wavelength of the illuminating radiation. If, however, two-electron pulses are caused by a simple overlap of single-electron ones, there should be no wavelength dependence of the pulse amplitude distribution because variation of the photocathode quantum efficiency only leads to a change in the probability of a single-electron pulse, while two-electron pulses emerge due to single-electron ones. If, while varying the wavelength, the single-electron pulse counting rate is kept constant, due to a corresponding variation of the illuminating light intensity, the count characteristic should apparently remain the same. This is exactly what is observed in experiment for a multialkali photocathode. The shape of the PMT output pulse amplitude distribution practically does not depend on the wavelength of the quasi-monochromatic light illuminating the photocathode (we have studied the wavelength interval from 600 to 1000 nm).

The results obtained for the multialkali photocathode can be summarised in the following way:

(i) Illumination of the photocathode by biphoton light led to a noticeable increase in the number of large-amplitude pulses at the PMT output, in a qualitative agreement with the results of Ref. [2]. No anomalously high number of two-electron pulses has been observed.

(ii) The number of two-electron pulses with the stretched leading edge was the same under illuminating the photocathode by biphoton light and by the light of gas plasma discharge. The counting rate of such pulses was in full agreement with the counting rate of pairs of single-photon events during the time interval of 7.2 ns.

(iii) When the photocathode was illuminated by radiation of a thermal light source with a narrow spectrum and various coherence times, as well as by laser spatially-coherent radiation, no difference in the counting rate of pulses with the stretched leading edge compared to the case of broadband thermal light source (radiation of an incandescent lamp transmitted through a monochromator) has been discovered.

(iv) The shape of PMT output pulse amplitude distribution was almost independent of the wavelength of quasi-monochromatic light source illuminating the photocathode.

A natural explanation of the results obtained for the multialkali photocathode is, to our viewpoint, as follows:

(i) Nonclassical pulses arising when the PMT photocathode is illuminated by biphoton light do not differ in shape from single-electron ones, due to the extremely short photon correlation time.

(ii) Most of the pulses with the stretched leading edge are classical ones.

(iii) The effect of photon bunching did not manifest itself in our measurements with illuminating the photocathode by spatially-coherent radiation of thermal sources due to the extremely short coherence times ($\sim$0.5 ns) compared to the integration time characterising two-electron pulses with the stretched leading edge.

(iv) Nonclassical pulses, which always exist, in a small number, when the photocathode is illuminated by the radiation of a thermal source, have probabilities that do not differ considerably from the probabilities of classical two-electron pulses.

7. Experimental evidence in favour of the existence of nonclassical two-electron pulses (results of the study of a cesium – antimonide photocathode)

Consider now the main question of this work, namely, the question whether it is possible to detect photon pairs with a probability considerably exceeding the squared probability of a single photon detection. To elucidate the situation, it is natural to try to reproduce the results obtained for the multialkali photocathode illuminated by biphoton light, for the case of a Sb–Cs photocathode. This would enable a direct measurement of the Sb–Cs photocathode quantum efficiency. Unfortunately, we did not manage to do it so far because of the low sensitivity of the Sb–Cs photocathode in the red spectral range (650 nm) where we generated nonclassical light. In this connection, we have tested, by means of indirect measurements, the statement of Ref. [1] about the anomalously high probability of two-photon photoeffect for the Sb–Cs photocathode. Apparently, there is not much sense in experiments on two-electron pulses with the stretched leading edge for FEU-64, since, due to a large pulse rise time (15 ns), such pulses are all classical. Therefore, we concentrated our efforts on a detailed test of the dependence of the pulse amplitude distribution on the wavelength of the illuminating quasi-monochromatic radiation obtained by passing the light from a SI6-100 incandescent lamp through a DMR-4 monochromator.

Consider first some new facts relating to the measurement of the single-electron peak contrast (the ratio of the maximal single-electron pulse counting rate to the counting rate at the local minimum of the count characteristic) carried out in our earlier work [3]. It is known that a Cs_3Sb photocathode is made of the semiconductor, whose electronic band-gap energy is 1.6 eV and the work function is 0.45 eV [10]. If one marks the energies corresponding to the band gap and to the photoeffect red boundary in the dependence of the single-electron peak contrast on the quantum energy of the incident monochromatic radiation (Fig. 6), an interesting picture arises. The contrast of the

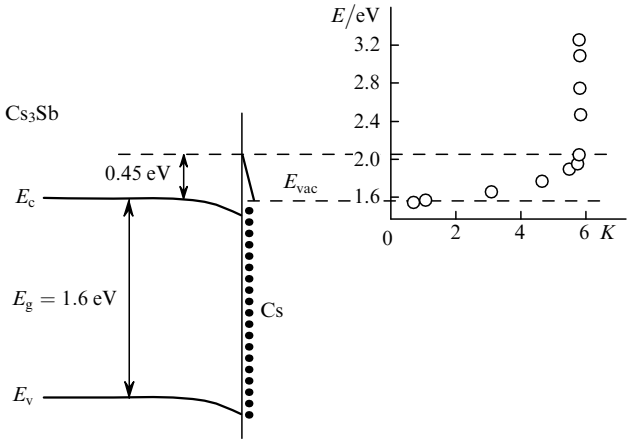

Figure 6. Energy bands of the Sb–Cs photocathode (near-surface bending of the bands and the potential barrier at the semiconductor–vacuum interface), left, and the dependence of the count characteristics of the single-electron peak contrast K on the quantum energy E of quasi-monochromatic radiation illuminating the photocathode, right; E_{vac} is the rest energy of a free electron.

single-electron peak starts to decrease when the quantum energy becomes less than the red photoeffect boundary; moreover, the single-electron peak virtually vanishes when the quantum energy is equal to the semiconductor band gap. A change in the single-electron peak contrast means, in fact, that the output PMT pulse amplitude distribution changes depending on the quantum energy of the radiation illuminating the photocathode. This was not observed for the multialkali photocathode.

We have performed a detailed study of the dependence of the FEU-64 PMT pulse amplitude distribution on the wavelength of the illuminating light, with the amplitude resolution much better than in Ref. [3] (using the same FEU-64 sample as in Ref. [3]). A higher amplitude resolution was achieved mainly due to using a higher-resolution MCA; this allowed us to work with signals of smaller amplitudes and to do without an Ortec-9302 amplifier, which had been used in Ref. [3]. (It should be noted that low amplification led to a decrease in the single-electron peak contrast.) It was discovered that the two-electron peak had a component depending linearly on the radiation intensity (Fig. 7). With a worse resolution, this two-electron 'shoulder' simply merges with the single-electron peak. If the light intensity increases, the amplitude of the two-electron 'shoulder' grows quadratically, and the shoulder becomes visible even for a moderate amplitude resolution. As a result, we can state that the paper by Artem'ev [1] mainly dealt with the linear (in the intensity) component of the two-electron peak, while Ref. [3] discussed the two-electron peak at higher intensities, with the quadratic contribution dominating. Pulse distributions obtained for relatively small illumination levels show that with the increase of the wavelength, the contribution of the single-electron peak gradually decreases and becomes comparable with the two-electron one.

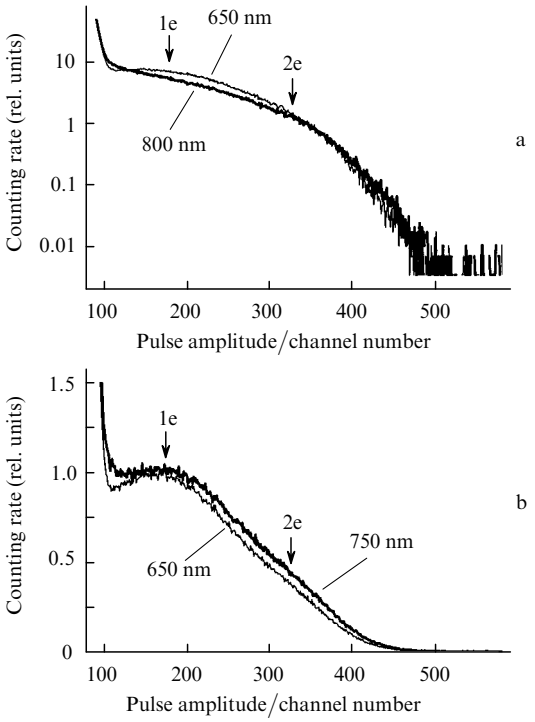

Figure 7. Count characteristics of a FEU-64 illuminated by quasi-monochromatic light with $\lambda = 650$ and 800 nm (a) and 650 and 750 nm (b). Curves in Fig. 7b are normalised to the amplitude of the single-electron peak. Arrows show the positions of the single-electron (1e) and two-electron (2e) peaks. The integral pulse counting rate is 10^3 pulse s^{-1} in both cases.

Fig. 7a shows, on the logarithmic scale, pulse amplitude distributions obtained for the same integral counting rate of 10^3 pulse s^{-1} and for quasi-monochromatic radiation with wavelengths 650 and 800 nm. One can see that as the wavelength increases, the counting rate in the vicinity of the single-electron peak drops faster than in the vicinity of the two-electron one, so that at $\lambda = 800$ nm the single-electron peak is not visible any more and the count characteristic becomes a monotonously decreasing curve. This can be observed in more detail if one compares both count characteristics in the linear scale. Fig. 7b shows such characteristics for two different wavelengths, normalised to the counting rate at the single-electron peak. The figure clearly shows the increase in the two-electron peak relative contribution and the related decrease in the single-electron peak contrast. A similar change in the shape of the FEU-64 count characteristic has been observed earlier in Ref. [11]. This two-electron peak is not two-photon at first sight since its height has a linear dependence on the intensity. This fact is important for the understanding of the photoeffect mechanism, and we will discuss it in detail below, while considering a model for the observed effects. However, there is a question that is principally important for the motivation of this work, namely: what is the contribution of definitely two-photon (i.e., quadratic in the intensity) photoeffect into the two-electron peak? Is this contribution indeed anomalously large?

The answer is given by Fig. 8, which shows how the pulse amplitude distribution changes due to the intensity increase for two different wavelengths. Distributions in Fig. 8 were obtained for the same integral pulse counting rates (10^3 and 23×10^3 pulse s^{-1}) and normalised to the amplitude of the single-electron peak. At a counting rate of 10^3 pulse s^{-1}, the quadratic contribution into the two-electron peak can be neglected; therefore, the integral pulse counting rates J_1 and J_2 at $\lambda = 650$ and 750 nm, respectively, can be written as

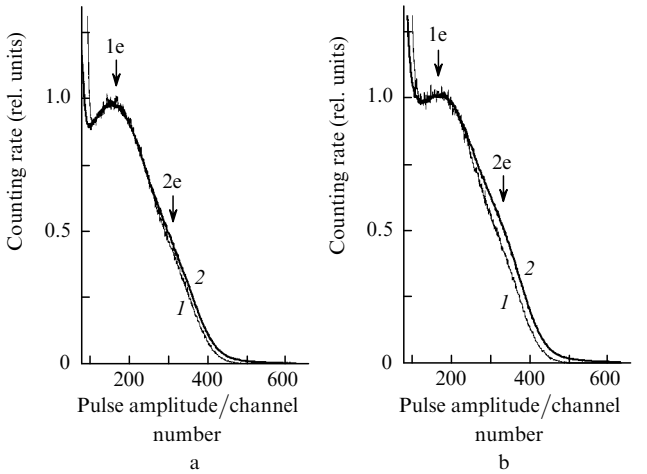

Figure 8. Count characteristics of a FEU-64 illuminated by quasi-monochromatic light with $\lambda = 650$ (a) and 750 nm (b) and the integral pulse counting rates of 10^3 (*1*) and 23×10^3 pulse s^{-1} (*2*). The curves are normalised to the amplitude of the single-electron peak. Arrows show the positions of the single-electron (1e) and two-electron (2e) peaks.

$$J_1 = (\eta_1 + \mu_1)\bar{I}_1,$$
$$J_2 = (\eta_2 + \mu_2)\bar{I}_2, \tag{3}$$

where $\bar{I}_1$ and $\bar{I}_2$ are mean intensities of the radiation at $\lambda = 650$ and 750 nm; η_1, η_2 and μ_1, μ_2 are single- and two-electron PMT efficiencies at the same wavelengths. Since the single-electron peak at $\lambda = 650$ nm contributes much more into the distribution,

$$\frac{\eta_1}{\eta_1 + \mu_1} > \frac{\eta_2}{\eta_2 + \mu_2}. \tag{4}$$

When the counting rate increases up to 23×10^3 pulse s^{-1}, we should take into account the quadratic in the intensity contribution into the two-electron peak. Then,

$$J_1' = (\eta_1 + \mu_1)\bar{I}_1' + \frac{(\eta_1 \bar{I}_1')^2}{2} T,$$
$$J_2' = (\eta_2 + \mu_2)\bar{I}_2' + \frac{(\eta_2 \bar{I}_2')^2}{2} T. \tag{5}$$

Here, T is the integration time of single-electron events. According to the experimental conditions, $J_1' = J_2' = J$. It follows from (4) that

$$\frac{\eta_1 \bar{I}_1'}{(\eta_1 + \mu_1)\bar{I}_1'} > \frac{\eta_2 \bar{I}_2'}{(\eta_2 + \mu_2)\bar{I}_2'}, \tag{6}$$

i.e.,

$$\frac{\eta_1 \bar{I}_1'}{J - (\eta_1 \bar{I}_1')^2 T/2} > \frac{\eta_2 \bar{I}_2'}{J - (\eta_2 \bar{I}_2')^2 T/2}. \tag{7}$$

One can see from Fig. 8 that the quadratic corrections to linear counting rates are rather small at $J = 23 \times 10^3$ pulse s^{-1}; hence, we obtain from Eqn (7) that

$$\eta_1 \bar{I}_1' > \eta_2 \bar{I}_2'. \tag{8}$$

Therefore, the quadratic contribution into Eqn (5) should be larger for $\lambda = 650$ nm than for $\lambda = 750$ nm, while the experiment shows an opposite behavior. Thus, we come to the conclusion that the two-photon efficiency is not equal to the squared single-photon one or, alternatively, the probability of a two-photon effect cannot be represented as a product of two single-photon effect probabilities.

8. A physical model providing the consistent explanation of the observed results

The experimental results presented above make us conclude that the anomalously high probability of two-electron pulses at the output of a photomultiplier is related not to the coherence of light illuminating the photocathode but to the material of the photocathode. The cesium–antimonide photocathode was one of the first photocathodes with large quantum efficiency and is rather well-studied at present. A significant feature of this photocathode and similar ones is an excess layer of cesium atoms applied to the surface in order to reduce the work function. It is a well-known fact that atoms adsorbed on pure surfaces of metals and semiconductors may considerably reduce the work function [12–15]. A decrease in the work function has been recently demonstrated for photocathodes based on n-type semiconductor materials such as GaN and GaAs [16–18]. This effect can be explained by the influence of strong near-surface electric field arising due to the transfer of cesium valence electrons into the semiconductor. Electron transfer leads to the creation of a space charge layer near the semiconductor surface. This results in the bending of the semiconductor energy bands in the near-surface area and in the accumulation of positive surface charge formed by charged donor centers of cesium. As a result, a strong electric field is formed near the surface, directed from the donor layer to the space charge layer, and this field accelerates electrons and helps them to exit the semiconductor [12, 13, 15].

The behaviour of electron energy near the surface is shown schematically in Fig. 6. A jump of this energy on the surface, leading to a potential barrier, is caused by short-range electric forces. One can see that applying cesium to the surface strongly changes the conditions for the electron exit into the vacuum. While in the case of a pure surface an electron has to overcome a step in the potential, in the case of adsorbed cesium an electron has to pass through a potential barrier. The spatial width of this barrier is 3–5 Å ([15], p. 429). Looking once again at the dependence of the single-electron peak contrast on the photon energy (Fig. 6), it is logical to assume that the barrier height is 0.45 eV, i.e., corresponds to the work function. Then it becomes clear why the single-electron peak contrast is independent of the photon energy at energies larger than 2 eV (over-barrier exit) and decreases at energies less than 2 eV (electron tunneling through the potential barrier). For the case of tunneling, the contribution of the two-electron peak increases compared to the single-electron one as the wavelength of the incident light increases. If the tunneling probability of an electron pair were equal to the square of a single electron tunneling probability, then the increase in the wavelength would not lead to any change in the relation between the single- and two-electron peak contributions, provided that the integral counting rate was kept constant. Hence, we should conclude that an electron pair can tunnel through a potential barrier with a probability considerably larger than the square of single-electron tunneling probability.

It is interesting that up to recently, no one considered the problem of two-electron tunneling, although the solution to the single-electron problem can be found in any textbook on quantum mechanics. The problem of two-electron tunneling was solved for the one-dimensional case and for the potential of an arbitrary shape in Ref. [19]. It turned out that under certain conditions, an electron pair can indeed tunnel through a potential barrier with a probability equal to the single-electron tunneling probability; besides, the probability of two-electron tunneling depends essentially on the total spin of the two-electron state. Note that the spin-orbit splitting of valence-band states may cause spin-polarised electrons to pass into the conduction band ([15], p. 442).

In order to obtain the self-consistent explanation of all given experimental results, we need to explain the linear in the intensity two-electron term in (3) and (5) and to interpret the disappearance of the two-electron 'shoulder' under

photocathode illumination by white light, which was observed in Ref. [1]. It should be noted here that the statements made in Ref. [1] on the absence of two-electron peak in the thermal noise of a PMT and in its count characteristic under white-light illumination cannot be confirmed by the experimental evidence given in Ref. [1]. Indeed, since the amplitude of PMT output pulses is strongly fluctuating, it is virtually impossible to rigorously distinguish single-electron pulses from dynode noise and two-electron pulses. The most complete description of the PMT output pulse amplitude distribution can be obtained based on the phenomenological model with a minimal number of fitting parameters [20]. Such an analysis was not carried out in Ref. [1]. The absence in the pulse distribution of a pronounced maximum or a 'shoulder', corresponding to the two-electron peak, means therefore only a considerable decrease of the two-peak contribution compared to the single-electron contribution. If it is not the radiation statistics that matters but the electrons tunneling through the potential barrier, then two-electron peak should be present in thermal noise as well, since it is not important whether we 'throw' electrons into the conduction band by light or due to thermal fluctuations.

Pulse amplitude distributions of thermal dark noise at two different temperatures are shown in Fig. 9. One can see that at a temperature of $t = 22\,^{\circ}\mathrm{C}$, there is a distinct bending point in the PMT count characteristic corresponding to the two-electron peak, although there is no pronounced two-electron peak. Note that for our FEU-64 sample, there is no pronounced single-electron peak in the dark thermal noise as well. The integral counting rate of pulses with amplitudes exceeding the dynode noise ones, was 2.3 pulse s^{-1} at the temperature $t = -25\,^{\circ}\mathrm{C}$ and 250 pulse s^{-1} at $t = +22\,^{\circ}\mathrm{C}$. If one estimates the probability of an electron pair tunneling during a time of the order of one nanosecond (as the squared probability of single-electron tunneling), then there is a clear discrepancy with the experimental data. Indeed, the average number of photocounts in 3 ns (the electron time of flight to the first dynode) is 7.5×10^{-7}. The square of this value, 5.6×10^{-13}, gives the mean counting rate of two-electron pulses 1.9×10^{-4} pulse s^{-1}, i.e., during the whole observation time (1 hour) there should be 6.7×10^{-1} such pulses, which is by several orders of magnitude less than their actual number. The actual counting rate of two-electron pulses, as we have mentioned before, is rather difficult to find under strong overlap of single- and two-electron peaks; but if it is estimated as the integral counting rate of pulses with amplitudes exceeding the amplitude of the two-electron peak maximum, we will get 3.7 pulse s^{-1}, which by four orders of magnitude exceeds the above estimate for the counting rate of quadratic events.

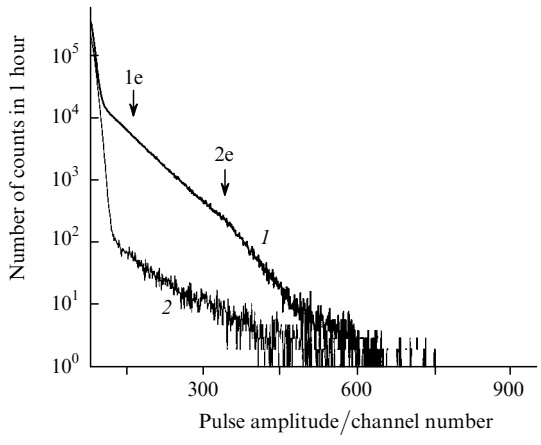

Figure 9. Dark count characteristics of a FEU-64 at the temperatures of $t = 22\,^{\circ}\mathrm{C}$ (*1*) and $-25\,^{\circ}\mathrm{C}$ (*2*). Arrows show the positions of the single-electron (1e) and two-electron (2e) peaks.

The reason for the two-electron 'shoulder' to disappear under white-light illumination can be found in the above-discussed spectral dependence of the PMT pulse amplitude distribution. Since white light contains all wavelengths, the resulting PMT pulse amplitude distribution should be a sum of all distributions obtained for the photocathode illuminated by monochromatic light, with the corresponding weighting coefficients. The two-electron peak is noticeable in the pulse amplitude distribution only for wavelengths within a certain interval of the red spectral range, where the PMT sensitivity is small; therefore, the single-electron peak will always dominate in the distribution under white-light illumination. In other words, if one compares two pulse amplitude distributions, the first one obtained for white-light illumination of the photocathode and the other one, under its illumination by monochromatic red light, the relative contribution of the two-electron peak will be less in the first case, in full agreement with the results of Ref. [1].

Another feature that becomes more important at higher light intensities is the time stability of light sources. The intensity of light emitted by the incandescent lamp used in Ref. [1] for obtaining white light, as a rule, is much more stable in time than the intensity of the PRK-2 bulb used for obtaining quasi-monochromatic radiation. It means that at equal mean intensities, the mean square of the intensity and hence the two-electron peak contribution, which is quadratic in the intensity, is larger in the case of quasi-monochromatic radiation than in the case of the incandescent lamp.

The situation with the linear in the intensity term in (5) is much more complicated. This term cannot be represented as resulting from a single-photon elementary process, i.e., absorption of a single photon and ejection of two electrons, since this is forbidden by energy conservation law. On the other hand, if the corresponding elementary process is a two-photon one, it should, at first sight, lead to a quadratic dependence of the counting rate on the light intensity. Indeed, let us be unable to resolve the absorption times of an electron within some short time τ_{in}. The probability P_2 of absorbing a photon pair should be then proportional to the squared integral intensity of light I during this time interval,

$$P_2(\tau_{\mathrm{in}}) = \left\langle \frac{\beta}{(\hbar\omega)^2} \int_t^{t+\tau_{\mathrm{in}}} I(t)I(t')\mathrm{d}t\mathrm{d}t' \right\rangle. \qquad (9)$$

Here, β is the two-photon efficiency of the photocathode and ω is the frequency of light, which is assumed to be monochromatic. The standard way to calculate expression (9) in terms of the normalised intensity correlation function $g_2(t, t')$ is based on the assumption that the angle brackets corresponding to ensemble averaging can be taken into the integral,

$$P_2(\tau_{\mathrm{in}}) = \frac{\beta}{(\hbar\omega)^2} \int_t^{t+\tau_{\mathrm{in}}} \langle I(t)I(t')\rangle \mathrm{d}t\mathrm{d}t' =$$

$$= \frac{\beta}{(\hbar\omega)^2}\bar{I}^2 \int_t^{t+\tau_{in}} g_2(t,t')\mathrm{d}t\mathrm{d}t'. \quad (10)$$

One can see that the probability of two-photon photoeffect will be in this case indeed a quadratic function of the mean intensity of light $\bar{I}$. However, if we cannot distinguish between the time moments t and t' due to the quantum nature of our measurements, (9) can be considered as the second moment of the integral intensity probability distribution,

$$S(t,\tau_{in}) = \frac{1}{\hbar\omega}\int_t^{t+\tau_{in}} I(t)\mathrm{d}t,$$

which in the case of a stationary process will only depend on τ_{in},

$$P_2(\tau_{in}) = \beta\langle S^2(t,\tau_{in})\rangle. \quad (11)$$

If $S(\tau_{in})$ is treated as the probability to discover a photon within the time interval τ_{in}, which is the usual assumption in the derivation of the semiclassical Mandel formula, forming the basis of the photodetection theory, then it apparently takes a continuous set of values. The probability distribution for $S(\tau_{in})$ in the case of a thermal light source with a Lorentzian spectrum has been found in Ref. [21], for an arbitrary relation between τ_{in} and the inverse spectral width. Using this distribution, one can find $\langle S^2(t,\tau_{in})\rangle$, which is especially easy in the limiting cases of time intervals τ_{in} that are large or small compared to the inverse spectral width. As a result of such calculation, one obtains again that the probability $P_2(\tau_{in})$ should depend on the mean intensity quadratically.

However, there exists another way of calculating $\langle S^2(t,\tau_{in})\rangle$, based not on the semiclassical photodetection theory but directly on thermodynamics. Indeed, at sufficiently small τ_{in} one can write $(\hbar\omega)^2\langle S^2(t,\tau_{in})\rangle = \langle E^2_{\Delta\omega}\rangle$, where $\langle E^2_{\Delta\omega}\rangle$ is the mean square energy of quasi-monochromatic thermal radiation. This value may be found thermodynamically without any additional assumptions on photon detection probability. It was first calculated by Einstein in 1909 when considering energy fluctuations of the blackbody radiation within a given frequency range $\Delta\omega$ [22],

$$\langle E^2_{\Delta\omega}\rangle = \hbar\omega\langle E_{\Delta\omega}\rangle + \left(1 + \frac{\pi^2 c^3}{V\omega^2\Delta\omega}\right)\langle E_{\Delta\omega}\rangle^2. \quad (12)$$

Here, V is the quantisation volume and c is the speed of light. It is important that the energy of thermal radiation within the quantisation volume (and hence its square) can only take integer values at the moment of the measurement, although its mean values certainly do not have to be integer. Since the blackbody radiation is in thermodynamic equilibrium with the material of the cavity containing this radiation, one can state that the mean square of the energy emitted and absorbed by the cavity within time $V^{1/3}/c$ is also given by Eqn (12), which contains not only a quadratic term in the intensity of light but also a linear one. At $\hbar \to 0$, the linear term disappears and Eqn (12) turns into an expression similar to the semiclassical formula (10). Thus, we can suppose that the observed two-electron peak whose amplitude has a linear dependence on the intensity is a two-photon one.

Note that there still exists no consistently quantum theory of photodetection. The problem of time measurements in quantum mechanics is being actively discussed [23] but is still far from being solved. When detecting a two-photon pulse, we do not detect each photon separately and can therefore expect that the well-known semiclassical photodetection theory, where the probability of multi-photon absorption is expressed in terms of single-photon absorption probabilities, will be non-applicable in this case. Moreover, if one sums up the probability amplitudes of separate photons present on the photocathode, as it is commonly done in quantum mechanics, and then finds the probability of discovering a photon pair by taking the square of this sum, the result will be namely proportional to the intensity of light. Of course, all these considerations are qualitative and cannot be considered as proofs. The final solution to the problem can be only found with the help of a consistent quantum-mechanical approach to the photodetection process, without using semiclassical recipes of probability calculation.

9. Conclusions

Thus, we have studied two-electron pulses in photomultipliers and demonstrated a principal possibility of detecting photon pairs with probabilities considerably exceeding the squared probability of single-photon registration. A model of observed effects is suggested, based on two-electron tunneling of photoelectrons through the potential barrier at the boundary between the photocathode and the vacuum. A method of detecting two-electron pulses with the stretched leading edge is described, which is of interest for single-channel studies of photocount statistics.

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Appendix

Consider the process of detecting pairs of electron pulses. The probability of detecting the first pulse at the instant t during the interval Δt is

$$p_1(t) = \eta I(t)\Delta t. \quad (A1)$$

This pulse triggers a TAC, so that the second pulse can be detected only after the time $t+\tau$, where τ is the TAC preparation time, and till the time $t+\tau+T$, where T is the time sweep range of the TAC. The time τ consists of the time τ needed for the TAC to prepare for the measurement

(5 ns) and the time delay at the 'start' input of the TAC. Thus, the probability to detect a pair of pulses at the time when the TAC is ready, is

$$p_2(t+\tau) = \eta^2 I(t)\Delta t I(t+\tau)\Delta t. \quad \text{(A2)}$$

In order to calculate the probability that the second pulse appears within the time sweep range of the TAC, it is necessary to take into account the probability that there are no pulses before this moment [24]. For instance, the probability to detect the second pulse at time $t+\tau+\Delta t$ is

$$p_2(t+\tau+\Delta t) = \eta I(t)\Delta t[1-\eta I(t+\tau)\Delta t]\eta I(t+\tau+\Delta t)\Delta t, \quad \text{(A3)}$$

and at time $t+\tau+N\Delta t$,

$$p_2(t+\tau+N\Delta t) = \eta^2 I(t)\Delta t \prod_{n=0}^{N-1}[1-\eta I(t+\tau+n\Delta t)\Delta t]$$

$$\times I(t+\tau+N\Delta t)\Delta t. \quad \text{(A4)}$$

The total probability to detect a pair during the sweep range $T = M\Delta t$ is

$$P_2(t,T) = \eta^2 I(t)\Delta t \sum_{N=0}^{M}\prod_{n=0}^{N-1}[1-\eta I(t+\tau+n\Delta t)\Delta t]$$

$$\times I(t+\tau+N\Delta t)\Delta t. \quad \text{(A5)}$$

If the intensity of light is not very large, so that $\eta I(t+\tau+n\Delta t)\Delta t \approx 0$, this probability can be represented as an integral

$$P_2(t,T) = \eta^2 I(t)\Delta t \int_{t+\tau}^{t+\tau+T} I(t+\tau+\xi)\mathrm{d}\xi. \quad \text{(A6)}$$

Averaging over time t leads, under the assumption that the light is stationary, to the expression

$$P_2(T) = \langle P_2(t,T)\rangle = \eta^2\Delta t \int_{t+\tau}^{t+\tau+T}\langle I(t)I(t+\tau+\xi)\rangle\mathrm{d}\xi$$

$$= \eta^2\Delta t\int_{\tau}^{\tau+T} G_2(\tau+\xi)\mathrm{d}\xi, \quad \text{(A7)}$$

where $G_2(\tau)$ is the intensity correlation function. For comparison with the experiment, it is convenient to use the mean pair counting rate, which, under neglecting the TAC dead time, is given by the expression

$$R_2(T) = P_2(T)/\Delta t = \eta^2\bar{I}^2\int_{\tau}^{\tau+T} g_2(\tau+\xi)\mathrm{d}\xi$$

$$= r^2\int_{\tau}^{\tau+T} g_2(\tau+\xi)\mathrm{d}\xi, \quad \text{(A8)}$$

where $g_2(\tau)$ is the normalised intensity correlation function. If $\tau, T \gg \tau_c$ (τ_c is the coherence time), then $g_2(\tau+\xi) = 1$, and the counting rate is simply

$$R_2(T) = \eta^2\bar{I}^2 T = r^2 T. \quad \text{(A9)}$$

Let us now apply the obtained formulas to the case of detecting two-electron pulses with the stretched leading edge. The above-given calculation scheme is applicable in the case where only a very small photocathode region is illuminated coherently, and one can neglect the difference between times of flight for electrons emitted by its different parts. Under this assumption, for obtaining a pulse with the stretched leading edge it is necessary that the difference between the exit times of electrons leaving the photocathode is comparable with the rise time τ_0 of the PMT single-electron pulse. All above considerations are valid in this case; also, the time T should coincide, up to the order of magnitude, with τ_0. Then, Eqn (A8) indicates that the measured counting rate of pulses with the stretched leading edge will be proportional to the integral of the correlation function with the lower integration limit $\tau = \tau_0$.

If the photocathode is illuminated by light with small coherence radius, the information about the statistical properties of light is rapidly lost with increasing the number of modes, due to a large number of uncorrelated photon pairs belonging to different modes. As a result, in the case of a large mode number, the counting rate of pulses with the stretched leading edge is given by Eqn (A9) regardless of the thermal source coherence time. Thus, if the photocathode is illuminated by light with small coherence time, the dependence of R_2 on r for classical two-electron pulses should be quadratic with the coefficient given by the integration time T; under coherent illumination, this coefficient slightly increases.

References

1. Artem'ev V.V. *Radiotekhn. Elektron.*, **9**, 756 (1964); **13**, 316 (1968); Artem'ev V.V., Gus'kov L.N., Mikhailov V.N. *Pribor. Tekhn. Exper.*, (4), 68 (1966).
2. Klyshko D.N., Penin A.N. *Usp. Fiz. Nauk.*, **152**, 653 (1987).
3. Lebedev M.V., Misochko O.V., Ainbund M.R. *Zh. Eksp. Teor. Fiz.*, **120**, 1351 (2001).
4. Lissandrin F., Saleh B.E.A., Sergienko A.V., Teich M.C. *Phys. Rev. B*, **69**, 165317 (2004).
5. Mandel L. *Proc. Phys. Soc.*, **74**, 223 (1959).
6. Morgan B.L., Mandel L. *Phys. Rev. Lett.*, **16**, 1012 (1966).
7. Becher C. et al. *Phys. Rev. B*, **63**, 121312(R) (2001).
8. Kiraz A. et al. *Phys. Rev. B*, **65**, 161303(R) (2002).
9. Kiraz A. et al. *Phys. Rev. A*, **69**, 032305 (2004).
10. Grigor'ev I.S., Meilikhov E.Z. (Eds) *Fizicheskie velichiny. Spravochnik* (Handbook of Physical Quantities) (Moscow: Energoatomizdat, 1991) p. 577.
11. Vetokhin S.S., Reznokov I.V., Vereshchagin V.L. *Prib. Tekhn. Eksper.*, (1), 166 (1974).
12. Soboleva N.A. *Usp. Fiz. Nauk.*, **111**, 331 (1973).
13. Berkovskii A.G., Gavanin V.A., Zaidel' I.N. *Vakuumnye fotoelektronnye pribory* (Vacuum Photoelectronic Devices) (Moscow: Energiya, 1976) p. 22.
14. Cardona M., Ley L. (Eds) *Photoemission in Solids I.* (Berlin – Heidelberg – New York: Springer-Verlag, 1978).
15. Lüth H. *Surfaces and Interfaces of Solids* (Berlin – Heidelberg – New York: Springer-Verlag, 1993).
16. Benemanskaya G.V., Daineka D.V., Frank-Kamenetskaya G.E. *Surface Sci.*, **523**, 211 (2003).
17. Benemanskaya G.V. et al. *Appl. Phys. Lett.*, **85**, 1365 (2004).

18. Benemanskaya G.V., Frank-Kamenetskaya G.E., Shmidt N.M., Dunaevskii M.S. *Zh. Eksp. Teor. Fiz.*, **130**, 506 (2006).
19. Hassler F., Suslov M.V., Graf G.M., Lebedev M.V., Lesovik G.B., Blatter G. cond-mat: arXiv:0802.0143v1.
20. Dossi R., Ianni A., Ranucci G., Smirnov O.Yu. *Nucl. Instrum. Meth. Phys. Res. A*, **451**, 623 (2000).
21. Jakeman E., Pike E.R. *J. Phys. A* (*Proc. Phys. Soc.*), **1**, 128 (1968).
22. Landau L.D., Lifshitz E.M. *Statistical Physics* (Oxford: Pergamon Press, 1980; Moscow: Nauka 1995) Pt. 1, p. 388.
23. Muga J.G., Sala Mayato R., Egusqiza I.L. (Eds) *Time in Quantum Mechanics* (Berlin: Springer-Verlag, 2002).
24. Davidson F., Mandel L. *J. Appl. Phys.*, **39**, 62 (1968).