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Photon statistics for the synchrotron radiation from electrons in a storage ring

I. S. Guk, N. N. Naugol'nyĭ, and A. S. Tarasenko

Physiotechnical Institute, Ukrainian Academy of Sciences
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Photon statistics have been investigated for the synchrotron radiation emitted by a bunch of relativistic electrons. Expressions are obtained for the variance of the photon number and the field density operator. The experimental method is described and experimental data confirming the theoretical analysis are reproduced.

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1. INTRODUCTION

The solution of a series of new applied problems has recently led to a much greater interest in the statistical properties of electromagnetic fields of different origin. The theory of coherence, developed by many authors, has led not only to a successful description, but also to the prediction, of a number of new properties for the two main classical sources, namely, thermal and coherent sources.[1] There is considerable interest in the statistics of synchrotron-radiation photons emitted by a bunch of relativistic charged particles because a source of this kind cannot be assigned to either of the above two classes. Moreover, detailed studies of the properties of synchrotron radiation are desirable in view of the increasingly extensive application of this radiation as a practical tool in physics generally.[2]

2. PHOTON STATISTICS AND THE FIELD-DENSITY OPERATOR FOR SYNCHROTRON RADIATION

Since synchrotron radiation losses per orbit in the storage ring of the Physico-technical Institute of the Ukrainian Academy of Sciences[3] are negligible in comparison with the energy of the radiating electron ($E = 100$ MeV), the recoil experienced by the electron can be neglected in the calculation of the synchrotron-radiation parameters. In this approximation, a relativistic electron can be regarded as a classical current, and Glauber has shown[4] that its radiation field is in a coherent state and is described by a set of complex amplitudes $\{\alpha_{l1}\}$.

The emission probability amplitude $\alpha_{l1}(t)$ for the l-th harmonic of orbital frequency ω and a charge e moving on a circular orbit of radius R is given by[5]

$$\alpha_{l1}(t)=\frac{\pi^{1/2}ev\varepsilon_{\perp}^{*}}{(2\omega_l\hbar)^{1/2}}\sum_{p=-\infty}^{\infty}\frac{1}{p\omega+\omega_l}\exp\left[i\left(\frac{3}{2}\pi(p-1)-p\varphi_l+p\psi\right)\right]\times J(p)\exp[i(p\omega+\omega_l)t], \quad (1)$$

where $\varepsilon_{\perp}$ is the projection of the polarization vector onto the plane of the orbit, $\varphi_l=\arctan(k_y/k_x)$, ψ is the initial phase of the particle, $\omega_l\simeq\omega l$, $v=\omega R$,

$$J(p)=J_{p-l}(Rk_{\perp})\exp[i(\varphi_l+\Delta_l)]+J_{p+l}(Rk_{\perp})\exp[-i(\varphi_l+\Delta_l)];$$

J_p is the Bessel function, $\Delta l=\arctan(\varepsilon_x^*/\varepsilon_y^*)$, and $k_{\perp}$ is the projection of the wave vector onto the plane of the orbit.

Consider the radiation emitted by a set of N noninteracting charges. For this ensemble,

$$\alpha_{lN}(t)=\sum_{\nu=1}^{N}\alpha_{l\nu}(t). \quad (2)$$

Under these conditions, $\langle |\alpha_{1N}|^2\rangle \equiv \langle nt\rangle$ is the number of photons emitted by the system within the time interval $(0, t)$, averaged over the corresponding distribution. It is important to note that this averaging procedure is purely quantum-mechanical. From (2), we have

$$\langle nt\rangle = \langle n_1 t\rangle \left\{N + \left\langle \sum_{\nu\neq\nu'} \exp[il(\psi_\nu - \psi_{\nu'})]\right\rangle\right\}, \tag{3}$$

where

$$\langle n_1 t\rangle = \frac{c^2e^2\beta^2l^2}{2\pi R^2} |\beta J_l'(\beta l \sin\theta)\varepsilon_1 - i\varepsilon_2 J_l(\beta l\sin\theta)\operatorname{ctg}\theta|^2\, do$$

is the shot photon intensity within a solid angle element do, $\beta = v/c$, and ε_1 and ε_2 are the components of the polarization vector.

The expression given by (3) can be written in the form

$$\langle nt\rangle = \langle n_1 t\rangle N\{1+(N-1)f\}, \tag{4}$$

where f is the formfactor, the explicit form of which is determined by the specific distribution of particles in the bunch.

The variance σ^2 of the number of photons is related to the mean value $\langle nt\rangle$ and the semi-invariant $\langle |\alpha|^4\rangle$ by the following expression[1a]:

$$\sigma^2 = \langle |\alpha|^4\rangle + \langle nt\rangle - \langle nt\rangle^2. \tag{5}$$

After averaging, we have from (1) and (2)

$$\langle |\alpha|^4\rangle = \langle n_1 t\rangle^2 N\{2N-1+4(N-1)^2 f + (N-1)\cdot(N^2-3N+3)f^2\}. \tag{6}$$

Using (4) and (6), we obtain the following final expression for the variance of the number of photons:

$$\sigma^2 = \langle nt\rangle + \langle nt\rangle^2 - \langle n_1 t\rangle^2 N\{1+4(N-1)f + (N^3-4N+3)f^2\}. \tag{7}$$

When $f = 0$ (total mutual incoherence of the particles), we find that $\sigma^2 = \langle nt\rangle + \langle n_1 t\rangle^2 N(N-1)$ and, when $N \to \infty$, which is the condition for the asymptotic approximation to the perfectly random type of excitation, we have $\sigma^2 = \langle nt\rangle + \langle nt\rangle^2$. When $f = 1$ (complete coherence), the result is $\sigma^2 = \langle nt\rangle$, i.e., the variance corresponds to the Poisson distribution of the number of photons.

From (4) and (7), it follows that, for equal synchrotron and thermal radiation intensities, the variance difference is

$$\delta = -\frac{\langle nt\rangle^2}{N}\left\{1 + \frac{2(N-1)f + (N^3-N^2-2N+2)f^2}{[1+(N-1)f]^2}\right\}, \tag{8}$$

showing an enhanced level of correlation in synchrotron radiation as compared with thermal radiation, and the fact that, for a constant intensity, the degree of correlation of synchrotron radiation increases with increasing number of particles.

We now return to (4) and (7) and substitute $\langle nt\rangle_T = \langle n_1 t\rangle N(1-f)$ and $\langle nt\rangle_C = \langle n_1 t\rangle N^2 f$. Equation (4) can then be written in the form

$$\langle nt\rangle = \langle nt\rangle_T + \langle nt\rangle_C.$$

Instead of (7), we obtain

$$\sigma^2 = \langle nt\rangle + \langle nt\rangle^2 - \langle nt\rangle_C^2 - \langle n_1 t\rangle^2 N\{1+4(N-1)f - (4N-3)f^2\}. \tag{9}$$

The first three terms in this equation represent the variance of a mixture of thermal and coherent fields. When $\langle n_1 t\rangle^2 N \ll 1$, Eq. (9) shows that the synchrotron radiation from a relativistic bunch is satisfactorily described by a simple mixture of thermal and coherent components. This state of the field corresponds to the density operator[6]

$$\hat\rho = \frac{1}{\pi\langle n\rangle_T}\int \exp\left\{-\frac{|\alpha-\zeta|^2}{\langle n\rangle_T}\right\} |\alpha\rangle\langle\alpha| d^2\alpha, \tag{10}$$

in which the integral is evaluated over the entire complex plane of α: $d^2\alpha = d(\mathrm{re}\,\alpha)\, d(\mathrm{im}\,\alpha)$ and ζ is the probability amplitude for the coherent signal.

3. STATISTICAL PROPERTIES OF A RADIATION FIELD AND THE PHOTOMULTIPLIER PULSE-HEIGHT SPECTRUM

In experiments concerned with the statistical properties of optical fields, photons are usually recorded individually by photomultipliers. Let us consider the pulse-height spectrum at the output of a photomultiplier when the photocathode receiving the radiation field consists of a large number of noninteracting atoms.

We shall suppose that we have a single electron distribution $P(1, m)$ when the distribution is the result of the ejection of a single photoelectron from the photocathode in a time interval $[0, \tau]$, where τ is the resolution time of the multiplying system. The distribution will be regarded as an n-electron distribution if it is the result of the ejection of n photoelectrons.

Since the probability of detection of n photons in a fixed interval of time is independent of the choice of origin of the time interval, we can take the instant of photon detection as $t = 0$. The probability that the photomultiplier system will produce a pulse consisting of m electrons is

$$P(m) = \sum_{n=0}^{\infty} P(n, \tau) P(n+1, m), \tag{11}$$

where

$$P(n, \tau) = \mathrm{Sp}\left\{\hat\rho : \frac{(\alpha_0 \tau a^+ a)^n}{n!} \exp(-\alpha_0 \tau a^+ a) :\right\}$$

is the probability that n photoelectrons will be released in a time τ (α_0 is a coefficient proportional to the quantum yield of the detector, a^+ and a are, respectively, the photon creation and annihilation operators at a given point in space),[1b] and $P(n+1, m)$ is the probability that the $n+1$ photoelectrons will produce m electrons at the photomultiplier output.

If we know $P(1, m)$, we can obtain the following expression for $P=(n+1, m)$:

$$P(n+1, m)=\sum\prod_{l=0}^{m} P(1, l)^{k_l} n!\left[\prod_l k_l!\right]^{-1} \delta(n, \Sigma l k_l)\,\delta(n+1, \Sigma k_l). \tag{12}$$

Finally, the resultant pulse-height distribution at the output of the photomultiplier, the photocathode of which intercepts the radiation field, is given by

$$P(m)=\sum_{n=0}^{\infty} \mathrm{Sp}\left\{\hat{\rho}: \frac{(\alpha_0 \tau a^+ a)^n}{n!} \exp(-\alpha_0 \tau a^+ a):\right\} \times \sum\prod_{l=0}^{m} P(1, l)^{k_l} n!\left[\prod_l k_l!\right]^{-1} \delta(m, \Sigma l k_l)\,\delta(n+1, \Sigma k_l). \tag{13}$$

It follows from (13) that the pulse-height spectrum at the photomultiplier output, when the photocathode interacts with the radiation field, is determined by the statistical properties of the radiation field and the spectrum of single-electron pulses.

Since the actual form of the distribution $P(1, m)$ is governed by a large number of factors[7,8] (electron collection coefficient for each stage, dynode multiplication coefficients, which can be different for different stages, and so on), we shall analyze the above method on the assumption that we are dealing with a perfect photomultiplier system, i.e., the electron collection coefficients of each stage are equal to unity, the multiplication coefficients for all the stages are equal, and the multiplication law is Poissonian for each stage. The distribution $P(1, m)$ can then be described by the following approximate expression[9]:

$$P(1, m) \approx \frac{e^{-p} p^k}{\Gamma(1+k)\, p^{N-1}}, \tag{14}$$

where p is the mean multiplication coefficient of a given stage, $k=m/p^{N-1}$, and N is the number of stages in the photomultiplier. Using (12) and (14), we have

$$P(n, m)=\frac{e^{-np}(np)^k}{p^{N-1}\Gamma(1+k)}. \tag{15}$$

Thus, if the pulse-height spectra are normalized to unity, and the photocathode is exposed to radiation fields described by density operators $\hat{\rho}_1$ and $\hat{\rho}_2$, the difference between the spectra is

$$\Delta P(k)=\sum_{n=1}^{\infty} \Delta\alpha(n-1, \tau) \frac{e^{-p} p^k}{\Gamma(1+k)}\left[e^{-(n-1)p} n^k-1\right] \frac{1}{p^{N-1}},$$

$$\Delta\alpha(n, \tau)=\mathrm{Tr}\left\{\hat{\rho}_1: \frac{(\alpha_0 \tau a^+ a)^n}{n!} \exp(-\alpha_0 \tau a^+ a):\right\} - \mathrm{Tr}\left\{\hat{\rho}_2: \frac{(\alpha_0 \tau a^+ a)^n}{n!} \exp(-\alpha_0 \tau a^+ a):\right\}. \tag{16}$$

To determine the coordinates of the characteristic points of the distribution $\Delta P(k)$ as a function of the parameters $\Delta\alpha(n-1, \tau)$, we can use the following expressions:

$$\Delta P(k)=\sum_{n=1}^{\infty} \Delta\alpha(n-1, \tau) \frac{e^{-p} p^k}{p^{N-1}\Gamma(k+1)}\left[e^{(n-1)p} n^k-1\right]=0, \tag{17a}$$

$$\frac{d}{dm}\Delta P(k)=\sum_{n=1}^{\infty} \Delta\alpha(n-1, \tau) \frac{e^{-p+k\ln p}}{p^{2(N-1)}\Gamma(1+k)}\left[\ln(np)\, e^{-(n-1)p+k\ln n} - \ln p+\psi(1+k)\left(1-e^{-(n-1)p+k\ln n}\right)\right]=0, \tag{17b}$$

where $\psi(1+k)$ is the logarithmic derivative of the gamma function.

In general, the solution of the transcendental equations (17a) and (17b) cannot be obtained in analytic form but, if the coordinates of the characteristic points are known from experiment, the corresponding parameters $\Delta\alpha(n-1, \tau)$ can be found by numerical methods.

Let us consider the form of the distribution $\Delta P(k)$ in the case of the example for which $\Delta\alpha(n-1, \tau)>0$ and n varies from 1 to 2. In this case, the solution of (17a) is $k=p/\ln 2$, and it is readily shown that:

(1) when $0 \leqslant k<p/\ln 2$, we have $\Delta P(k)<0$; when $p/\ln 2<k \leqslant \infty$, we have $\Delta P(k)>0$;

(2) when $k \gg p/\ln 2$ and $k \ll p/\ln 2$, we have $d\Delta P(k)/dm<0$; when $k \sim p/\ln 2$, we have $d\Delta P(k)/dm>0$.

Therefore, $\Delta P(k)$ is a sign-varying function with two extrema: $k_{\min}<p/\ln 2$ and $k_{\max}>p/\ln 2$.

4. INVESTIGATION OF THE STATISTICAL PROPERTIES OF OPTICAL FIELDS BY MEASUREMENT OF THE PULSE-HEIGHT SPECTRUM PRODUCED BY A PHOTOMULTIPLIER

To investigate the possibility of using the analysis of pulse-height spectra produced by a photomultiplier as a means of studying the statistical properties of optical fields with short coherence times (10^{-11}–10^{-12} sec), we carried out a series of experiments with thermal radiation sources of different degree of coherence.

The experiment is illustrated schematically in Fig. 1. Radiation from source 1, which was a stabilized hot-filament or gas-discharge lamp filled with neon, was focused by a long focal length lens L_1 on the entrance slit of the UM-2 monochromator 2. The radiation emerging from the exit slit of the monochromator was focused by the short focal length lens L_2 on the photocathode of the FÉU-79 photomultiplier 3, which was arranged to count single photons. Pulses from the photomultiplier were amplified by the spectrometric amplifier 3 and were subjected to pulse height analysis in the

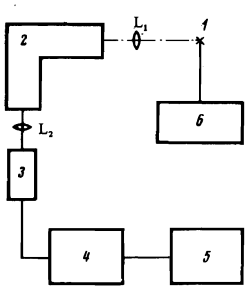

FIG. 1. Experimental arrangement for the investigation of the statistical properties of thermal radiation: 1—radiation source, L_1 and L_2 lenses; 2—monochromator, 3—FÉU-79 photomultiplier, 4—spectrometric amplifier, 5—pulse-height analyzer, 6—stabilized source of supplies.

analyzer 4. The resolving time T of the system was about 3×10^{-6} sec.

The equipment was first allowed to warm up for two hours. Since the maximum capacity of each analyzer channel was 64 000 pulses, the necessary precision was achieved by examining the pulse-height spectrum produced by the photomultiplier several times under the same conditions. The exposure time was chosen on the condition that there was no overload in any of the analyzer channels. At each exposure, we recorded the "signal + noise" spectrum and then for an equal interval of time we recorded the photomultiplier noise alone. To eliminate the effect of slow variations in the measuring-system parameters on the final result, we used different exposures in turn under different illumination conditions.

We recorded the pulse-height spectra produced by a photomultiplier whose photocathode was illuminated with the $\lambda = 5852$ Å neon line and white light of the same average wavelength with bandwidths $\Delta\lambda = 10$ Å and $\Delta\lambda = 400$ Å. The mean counting rate in all these cases was 5000 ± 10 sec^{-1}.

Since the geometry of the experiment was kept constant in all these measurements, it was assumed in the analysis of the spectra that the change in the probability that a photoelectron would produce a zero-amplitude pulse at the photomultiplier output was negligible when the conditions of illumination of the photocathode were varied.

Figures 2 and 3 show the difference between the suitably normalized pulse-height spectra obtained when the photocathode was exposed to white light with $\lambda = 5852$ Å, $\Delta\lambda = 10$ Å (Fig. 2) and $\Delta\lambda = 400$ Å (Fig. 3), and the neon line $\lambda = 5852$ Å, $\Delta\lambda < 1$ Å.

The quantity Δ plotted along the vertical axis is defined by

$$\Delta = (P_i - P_{i\,\mathrm{Ne}})/P_{\max \mathrm{Ne}},$$

where P_i is the probability of recording a pulse of

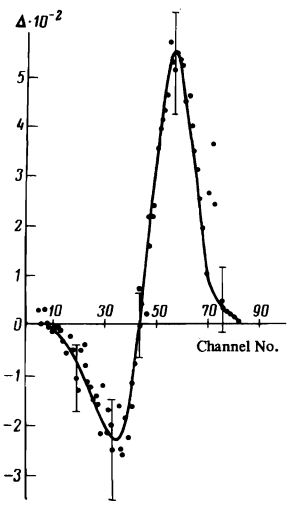

FIG. 2. Difference between normalized photomultiplier pulse-height spectra obtained with the cathode illuminated by white light with $\lambda = 5852$ Å, $\Delta\lambda = 10$ Å and the neon line $\lambda = 5852$ Å, $\Delta\lambda < 1$ Å.

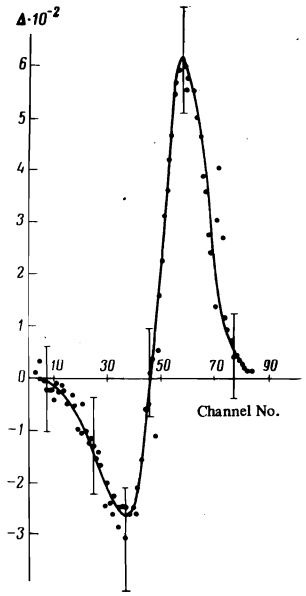

FIG. 3. Difference between normalized photomultiplier pulse-height spectra obtained with the cathode illuminated by white light with $\lambda = 5852$ Å, $\Delta\lambda = 400$ Å and the neon line $\lambda = 5852$ Å, $\Delta\lambda < 1$ Å.

height corresponding to the i-th channel of the analyzer when the photocathode is illuminated by a segment of the white-light spectrum, and $P_{i\,\mathrm{Ne}}$ is the probability of recording a pulse of the same amplitude when the photocathode is illuminated by the neon line; $P_{\max \mathrm{Ne}}$ is the maximum probability of recording a pulse of a particular amplitude when the photocathode is exposed to the neon line.

The dependence of Δ on pulse height (Figs. 2 and 3) is in accordance with the conclusions that can be drawn from (17), and indicates an enhanced content of multielectron components in the pulse-height spectra obtained when the photocathode is exposed to a rectangular spectrum with $\lambda = 5852$ Å, $\Delta\lambda_1 = 10$ Å, $\Delta\lambda_2 = 400$ Å, as compared with the spectrum due to the neon line $\lambda = 5852$ Å, $1\text{Å} > \Delta\lambda$.

It is well known that, if T is the duration of observations, and is much greater than the coherence time γ^{-1}, the probability of zero events (this quantity determines the percentage number of single-electron pulses in the pulse-height spectrum) for the rectangular and the Lorentz spectra is, respectively,[1c]

$$P_R(0,T) = (1+\mu_R)^{-\alpha} \qquad \alpha \equiv \frac{\bar{m}_R}{\mu_R},$$

$$P_L(0,T) = \exp\left\{-\frac{\bar{m}_L}{\mu_L}\left[(1+2\mu_L)^{1/2}-1\right]\right\},$$

where $\bar{m} = wT$, w is the mean counting rate, and $\mu = w/\gamma$.

When $\mu_R \ll \mu_L \ll 1$ and $\bar{m}_R = \bar{m}_L$, which was the case in our experiment, we find that $P_R(0,T) < P_L(0,T)$, i.e., the percentage contribution of multielectron pulses to the pulse-height spectrum is greater when the photocathode is illuminated by the rectangular white spectrum than when it is exposed to the Lorentz spectrum, which is in qualitative agreement with the results shown in Figs. 2 and 3.

Quantitative estimates for the experimental condi-

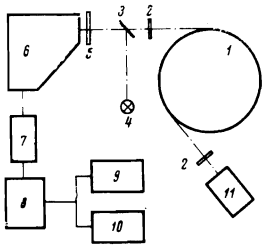

FIG. 4. Block diagram of the experiment on the statistics of synchrotron radiation.

tions with $\bar{m}_R = \bar{m}_L \sim 2.5\times10^{-2}$, $\mu_L \sim 5\times10^{-6}$, and $\mu_R \sim 5.5\times10^{-9}$ yield

$$\frac{P_R(m)-P_L(m)}{P_{L\max}}\Big|_{\max} \sim 10^{-7},$$

which is much less than the experimental result ($\sim 10^{-3}$).

Similar experimental results, i.e., substantially enhanced concentration (~ 2–10% of the total number of counts) in the spectrum of multielectron pulses, were reported previously in[10].

5. EXPERIMENTAL INVESTIGATION OF THE STATISTICAL PROPERTIES OF THE SYNCHROTRON RADIATION FIELD

The above method has a higher sensitivity than the coincidence method, and this enabled us to use it to investigate the statistical properties of the synchrotron radiation field. Figure 4 illustrates schematically the experimental arrangement.

Synchrotron radiation from the vacuum chamber of storage ring[3] 1 was extracted through the transparent window 2 and the semitransparent window 3, and, in some cases, through the neutral filter 5 as well. It was eventually intercepted by the entrance slit of the monochromator 6. The number of particles in the storage ring was measured by examining the synchrotron radiation with the photomultiplier 11, which was used as an integrating device and was calibrated for the amount of radiation per electron. The number of electrons could be determined to within about 10%. The source of the thermal radiation was a stabilized hot-filament lamp 4. The distance between the hot-filament lamp and the electron orbit and between the lamp and the entrance slit of the monochromator was the same. The radiation from the monochromator was focused onto the photocathode of the FÉU-79 photomultiplier 7 operating under single-electron conditions. Pulses from the photomultiplier were amplified by the spectrometric amplifier 8 and were counted by the scaling system 9 or were analyzed by the AI-128 pulse-height analyzer 10. The resolving time of the system was 3×10^{-6} sec.

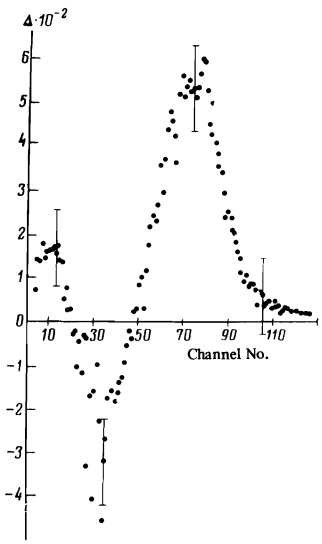

FIG. 5. Difference between normalized pulse-height spectra when the photocathode is exposed to thermal and synchrotron radiation.

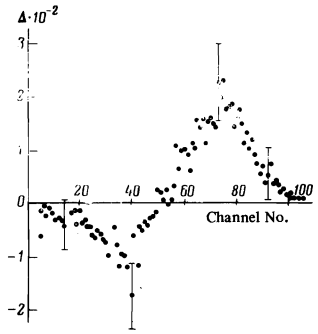

FIG. 6. Difference between normalized pulse-height spectra when the photocathode is exposed to synchrotron radiation due to a large ($\sim 1.2\times10^{10}$) and small ($\sim 1.2\times10^{6}$) number of radiating electrons.

Since the synchrotron radiation from electrons in the storage ring was in the form of a sequence of pulses, 3 nsec long and with a repetition period of ~ 20 nsec, whereas the radiation from a thermal source was continuous, this was taken into account in the comparison of the statistical properties of the two kinds of radiation. The monochromator was used to define a wavelength band with $\Delta\lambda = 20$ Å and $\lambda_0 = 5853$ Å. The mean counting rate for synchrotron-radiation photons was 3×10^{8} sec^{-1} for $N \approx 7\times10^{8}$ electrons, and was held constant during the exposure to within ± 5%.

Figure 5 shows the difference between the normalized pulse-height spectra from the photomultiplier, obtained when the photocathode was exposed to thermal radiation with $\Delta\lambda = 10$ Å and $\lambda_0 = 5852$ Å, and the synchrotron radiation of the same wavelength and the same band-width. The quantity Δ is defined by

$$\Delta = (P_{iT} - P_{is})/P_{\max s},$$

where P_{iT} is the probability of recording at the photomultiplier output a pulse of amplitude corresponding to the i-th analyzer channel when the photocathode is exposed to the thermal radiation, P_{is} is the same probability when the photocathode is illuminated by the synchrotron radiation, and $P_{\max s}$ is the maximum probability of detecting pulses of given amplitude when the photocathode is exposed to the synchrotron radiation.

The dependence of the statistical properties of synchrotron radiation on the number of radiating electrons was investigated for $N_1 = 1.2\times10^{10}$ and $N_3 = 1.2\times10^{6}$, where N is the number of electrons. The radiation intensity was equalized with the aid of neutral filters, keeping the geometry of the experiment unaltered. The counting rate was 5000 sec^{-1} in both cases. The measurements were performed in the same spectral region

as before. The difference between the pulse-height spectra for N_1 and N_2, normalized in the same way as before, is shown in Fig. 6.

The shape of the curve shown in Figs. 5 and 6 indicates that, for equal intensities and bandwidths, the synchrotron-radiation spectrum is less correlated than the thermal spectrum, and the degree of correlation of the synchrotron-radiation photons increases with increasing number of radiating particles.

6. CONCLUSIONS

The foregoing results lead to the following conclusions:

1. For equal mean intensities, thermal radiation has a higher degree of correlation than synchrotron radiation.

2. The degree of correlation of synchrotron radiation increases with increasing number of radiating electrons.

3. The efficiency of recording multiphoton pulses was found to be greater than predicted by the existing theory of photoelectric detection.

4. Analysis of the photomultiplier pulse-height spectra can be used to detect correlations in low-intensity fluxes with characteristic times of $\sim 10^{-11}$–10^{-12} sec.

5. Analysis of the pulse-height spectra produced by a photomultiplier can be effectively used to determine the statistical properties of an unknown source of radiation by comparing the resulting signal with the signal from a source with known statistical properties.

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Nonlinear quenching of the fluorescence of high-density localized electron excitations in molecular crystals

V. A. Benderskiĭ, V. Kh. Brikenshteĭn, M. A. Kozhushner, I. A. Kuznetsova, and P. G. Filippov

Institute of Chemical Physics, USSR Academy of Sciences, Moscow

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An investigation was made of the reduction in the quantum efficiency of the fluorescence of pyrene crystals (nonlinear quenching) observed when the optical pumping rate was increased in the range 10^{25}–10^{28} cm^{-3}·sec^{-1} at thermostat temperatures 4.2–300°K and the concentration of excitations reached 10^{-3}–10^{-2} of the density of molecules in a crystal. At 300°K this nonlinear quenching could be described by a model of bimolecular diffusion-controlled recombination. The excimer–excimer annihilation constant was found to be $(4\pm1)\times10^{-11}$ cm^3·sec^{-1}. Below 77°K, when the diffusion of excitations was negligible, the nonlinear quenching was due the dipole-dipole interaction of localized excitations. A model was developed on the basis of averaging of the kinetic equations for the random distribution and it was found that the nonlinear quenching could be described by introducing average self-consistent population. A good agreement was obtained between the experimental and theoretical dependences of the nonlinear quenching on the pumping rate and the effective radius (15–20 Å) was found for the dipole-dipole interaction between eximers.

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INTRODUCTION

Reduction in the quantum efficiency of the fluorescence of molecular crystals with rising optical pumping rate, called the nonlinear quenching, was discovered[1] and investigated in detail[2,3] in anthracene crystals. This quenching was attributed in [2,3] to the bimolecular interaction between excitons as a result of which one of the excitons is transferred to a higher vibronic state at the expense of the energy of a second exciton and this is