

Noise spectrum of spontaneous emission

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We have investigated the power spectrum for fluctuations in the spontaneous emission of mercury vapor ($\lambda=253.7$ nm). Predictions of the shape of the spectrum based on semiclassical and quantum theories are shown to be mutually exclusive. On the other hand, the experimental result is in agreement with the quantum-mechanical description of the elementary emission process. Analysis of fluctuations in the luminescence spectrum of mercury, excited by a noise-law modulated electron beam, has yielded information on the lifetime of the 6^1S_0 state of mercury.

1. INTRODUCTION

This paper is concerned with the fluctuation spectrum of the spontaneous radiation from an ensemble of non-interacting atoms. This problem is of interest, above all, as a very rare example of a sharp conflict between semiclassical and quantum mechanical descriptions of the elementary process of emission.

Any semiclassical analysis is essentially based on the classical description of the electromagnetic field and the quantum mechanical description of the quantum systems interacting with this radiation. This approach can be used in practically all cases to predict correctly the result of this kind of interaction. The semiclassical analysis is very attractive because of its ease of interpretation and clear logic. It has recently been particularly successful in the description of time-dependent resonance interactions (adiabatic inversion of populations, spin and photon echo, magnetic and optical nutations, and so on), parametric and nonlinear processes in optics, state interference phenomena, and also experiments on photon correlations. In all these cases, the rigorous approach demanding field quantization leads to identical results. It would appear that it is precisely these successes that have led to the popularity of the semiclassical methods. In recent years these methods, or certain modifications of the quantum theory made in the spirit of semiclassical ideas, have been used more or less successfully to consider such—at first sight purely quantum mechanical—questions as spontaneous emission, the photoelectric effect, resonance fluorescence, Lamb shift, and so on^[1-9] (most of these papers are reviewed in^[7] and^[10]). On the other hand, it has been said that radiation-field quantization is necessary only for the accurate calculation of radiation corrections. In this connection there is particular interest in the elementary situation where the semiclassical and quantum theories give, qualitatively, mutually exclusive predictions.¹⁾ In this paper we report an experimental study of the validity of the semiclassical approach to the decay kinetics of the excited states of atoms. In essence, we are concerned with the physical significance of the elementary process. Two alternative descriptions are compared:

1) The semiclassical description in which the decay of the excited state is treated in the spirit of the correspondence principle as the damping of an oscillator and is accompanied by the continuous emission of a real electromagnetic wave packet, where the amplitude of the oscillations and the damping constant are expressed in terms of the matrix elements for the corresponding transition; the subsequent interaction of the field with a quantum-mechanical system (photodetector) leads to a

discrete detection event which is interpreted as the detection of a photon.

2) The quantum-mechanical description in which the damped field assigned to the radiating atom determines only the probability amplitude for the detection of the photon and does not in itself have a physical meaning, i.e., we have the analog here of the wave-function field for an electron in an atom.

The results obtained in this paper are in agreement with the second description and show that the atomic emission process is discrete in itself, independently of whether the recording device is discrete or not. This is not a new conclusion but it would appear that the present investigation provides the first direct experimental demonstration of its validity.

2. SEMICLASSICAL ANALYSIS

Our description will be based on the following simple idea. If we consider a system of noninteracting atoms excited independently in accordance with the random law, then provided the decay kinetics is the same for all the atoms, one would expect that the intensity fluctuation spectrum due to the system will exhibit properties which will reflect the spectrum of the elementary emission process. This conclusion can readily be accepted in the case of a classical detector, i.e., a detector which can follow the variation in the field intensity in a continuous fashion. However, the situation is complicated if the discrete nature of the detector response has to be taken into account. It is clear that the discrete detector is basically incapable of following the decay of a single atom since, at best, it can react to such an event by the single action of emitting an electron which does not carry any information about the structure of the electromagnetic wave packet producing the emission.²⁾ However, in an experiment using an ensemble of atoms, the photodetector is subjected to the action of the resultant field, the fluctuations of which are readily seen to be connected with the law of decay of the atom and should lead to a characteristic grouping in time of the discrete photoemissive events. In particular, if the decay of the excited atoms occurs exponentially, one should be able to observe against the 'white' background due to shot noise (connected with the discrete nature of the photodetection process) the excess noise peak centered on zero frequency and having a width of the order of the natural width of the radiating level. The foregoing, relatively obvious prediction, based on the assumption of continuous emission in the elementary event, is confirmed by calculations whose main aim is to establish a quantitative measure for the required excess noise.

The photoelectric detection process in the semiclassical

sical approximation is described by a stochastic temporal sequence consisting of detection events for photoelectrons emitted from the photocathode under the action of the incident electromagnetic radiation.^[11] The electric signal at the output of an arbitrary linear circuit connected to the photodetector can be written in the form

$$U(t) = \sum_{k,i} u(t-t_k) n(\Delta_k t, \Delta_i s), \quad (1)$$

where $\{\Delta_k t\}$ is some sufficiently fine subdivision of the time of observation into elementary nonintersecting intervals containing the times $\{t_k\}$, $\{\Delta_i s\}$ is an analogous subdivision of the photocathode area, $n(\Delta_k t, \Delta_i s)$ is the number of photoelectrons appearing in the interval $\Delta_k t$ on the area $\Delta_i s$, and $u(t-t_0)$ is the response of the circuit to a single electron appearing at time t_0 (pulse characteristic of the circuit).

The stochastic properties of the signal $U(t)$ are determined by the statistics of the occupation numbers $\{n(\Delta_k t, \Delta_i s)\}$ which in turn is described by a bistochastic Poisson process.^[12] In particular, the joint probability distribution for the occupation numbers $n_{ki} = n(\Delta_k t, \Delta_i s)$ is

$$P(\{n_{ki}\}) = \prod_{k,i} \frac{\mu(\Delta_k t, \Delta_i s)^{n_{ki}}}{n_{ki}!} \exp[-\mu(\Delta_k t, \Delta_i s)], \quad (2)$$

where the positive quantities $\{\mu(\Delta_k t, \Delta_i s)\}$ describe the space-time dependence of the intensity of the photoelectron production process and, in general, are subject to a probability distribution. In the ensuing discussion we shall be interested only in the second moments of the distribution (2), which are necessary for the calculation of the correlation function for the signal (1):

$$\langle n(\Delta_k t, \Delta_i s) n(\Delta_j t, \Delta_m s) \rangle = \langle \mu(\Delta_k t, \Delta_i s) \mu(\Delta_j t, \Delta_m s) \rangle + \delta_{kj} \delta_{im} \langle \mu(\Delta_k t, \Delta_i s) \rangle, \quad (3)$$

where the angle brackets represent averaging over the corresponding distributions.

It follows from the semiclassical theory of photoelectric detection^[11] that the elementary intensity of the photoelectron current is given by

$$\mu(\Delta t, \Delta s) = q I(t, r) \Delta t \Delta s, \quad (4)$$

where q is the quantum efficiency of the photodetector and $I(t, r)$ is the instantaneous intensity of the optical field at the point r at time t . Using (1), (3) and (4), we can readily show that the correlation function for the electrical signal is

$$\begin{aligned} K_U(t, t') &= \langle U(t) U(t') \rangle - \langle U(t) \rangle \langle U(t') \rangle = q^2 \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} u(t-t_1) u(t'-t_1) dt_1 dt_2 \\ &\quad \times K_F(t_1, t_2) + q \int_{-\infty}^{\infty} u(t-t_1) u(t'-t_1) dt_1 \langle F(t_1) \rangle, \\ K_F(t_1, t_2) &= \int_S \int_S [\langle I(t_1, r_1) I(t_2, r_2) \rangle - \langle I(t_1, r_1) \rangle \langle I(t_2, r_2) \rangle] ds_1 ds_2, \\ \langle F(t_1) \rangle &= \int_S \langle I(t_1, r) \rangle ds, \end{aligned} \quad (5)$$

where the integral sums generated by the subdivisions $\{\Delta_k t\}$ and $\{\Delta_i s\}$ are replaced by integrals by going to the limit in which the time of observation is assumed to be unbounded, and integration with respect to the space coordinates is extended over the entire surface area S of the photodetector.

If we suppose that the optical intensity fluctuations are stationary in time, we can consider the spectral

representation of the process $U(t)$ in the form

$$G_U(\omega) = |\tilde{u}(\omega)|^2 \{q^2 G_F(\omega) + q \langle F \rangle\}, \quad (6)$$

where G_U and G_F are the fluctuation power spectra [the Fourier transforms of the corresponding correlation functions $K_U(\tau)$ and $K_F(\tau)$]. $\langle F \rangle$ is the mean flux at the photodetector, and $\tilde{u}(\omega)$ is the frequency characteristic of the electron circuit. It is clear from (6) that the spectrum $G_U(\omega)$ is the product of the known frequency characteristic of the detection circuit and the intrinsic fluctuation spectrum of the photoemission process. The latter contains two parts. The term $q \langle F \rangle$ is a consequence of the fact that photoemission is discrete, i.e. it is the 'white' shot noise. It complicates the analysis of the second component of the photoemission noise, which is connected with fluctuations in the light field intensity, but does not in principle prevent this analysis. It is precisely this component that is of main interest.

The evaluation of $G_F(\omega)$ is given in Appendix 1. We now use the following model of emission by an excited atom:

(1) the atoms are excited independently of one another at definite (random) instants of time,

(2) the excited atom emits a field in the form of a continuous, damped, scalar, spherical wave,

(3) the parameters describing the fields emitted by the different atoms are random, statistically independent, and identically distributed,

(4) the total number of atoms excited in the volume V during the observation T [$N = N(T, V)$] is a random quantity, is independent of the parameters describing the fields due to the individual atoms, and is described by a distribution whose first moments are related by

$$\langle N^2 \rangle = \langle N \rangle^2 + \langle N \rangle.$$

Condition (1) imposes a restriction on the type of excitation. It is undoubtedly satisfied in the case of electronic excitation and also when the spectrum of the optical excitation is much broader than the natural width of the excited state. Condition (2), with certain minor modifications concerned with the shape of the emitted wave, is the basis for any semiclassical description. Condition (3) is satisfied provided the concentration of the atoms is not too high. Finally, condition (4) corresponds to the Poisson distribution and is definitely satisfied when the number of excited atoms is small in comparison with their total number.

The details of the calculations are given in Appendix 1. Here we merely quote the final expression which is valid for frequencies ω much smaller than the (Doppler) width of the spectral line emitted by the ensemble of atoms:

$$G_U(\omega) = |\tilde{u}(\omega)|^2 q \langle F \rangle \left[1 + q \delta + q \frac{\Omega a(\omega)}{4\pi} \right] \quad (7)$$

In this expression δ is the degeneracy parameter for the light field,^[11] which is numerically equal to the number of photons passing through the coherence area during the coherence time, and Ω is the solid angle within which the detector collects light from the source.

In accordance with (7), the photoemission fluctuation spectrum consists of three components. As noted above, the first term represents shot noise. The other two give the excess noise connected with the optical-field statis-

tics and is zero in the special case of a regular field (single frequency laser). More detailed analysis reveals that the second term describes the excess (wave) noise arising from the interference between radiation emitted by different atoms. This radiation was investigated by Hanbury Brown and by Twiss. It is usually very small: for nonlaser sources of light $\delta < 0.001$ ^[11] and $q\delta \ll 1$. Since, moreover, this component is constant over the spectrum in the region which we are considering, we shall not take it into account henceforth: its presence does not affect the detection of the third component of the power spectrum $|u(\omega)|^2 q^2 \langle F \rangle \Omega a(\omega)/4\pi$ in which we are interested. This component of the excess noise is considered for the first time. It appears when we take into account the characteristic decay kinetics of the excited atom. The function $a(\omega)$ is the normalized mean power spectrum of the elementary emission process. It is clear from (7) that the power carried by this particular excess-noise component depends on the quantum yield q of the detector and the collection factor $\Omega/4\pi$, and in the limit ($q\Omega/4\pi = 1$, $\omega = 0$) may approach the shot noise power. We note that the predicted new term in the noise spectrum is obtained at the same time as the other two known components, whose form in the quantum mechanical and semiclassical descriptions is the same. This gives us confidence in the belief that the new result is valid.

3. EXPERIMENT

The aim of the experiment was to try to detect the excess intensity fluctuations in the spontaneous emission which are predicted by the semiclassical theory and are governed by the spontaneous emission kinetics. We shall ignore interference phenomena^[13] which are unimportant in the detection of radiation in a large solid angle, and shall assume that the decay of the atoms from excited states occurs exponentially with a damping constant Γ . In this case, equation (7) has the following form for $q\delta \ll 1$:

$$G_v(\omega) = |\bar{u}(\omega)|^2 q \langle F \rangle \left(1 + q \frac{\Omega}{4\pi} \frac{\Gamma^2}{\Gamma^2 + \omega^2} \right). \quad (8)$$

The normalized graph of this function is shown in Fig. 1.

Standard spectroanalytic equipment is inconvenient for the experimental detection of the excess noise because the maximum excess noise exceeds the shot-noise level by only 1% (see below). It was therefore decided to carry out careful comparison between the noise power in two fixed frequency intervals one of which, $\Delta\omega_l$, corresponds to the low-frequency excess-noise plateau, and the other, $\Delta\omega_h$, includes the region in which the excess noise practically ceases to fall (Fig. 1). The width of these intervals must be as large as possible,^[14] but it is clear that it must satisfy the condition $\Delta\omega_l = \Delta\omega_h < \Gamma$.

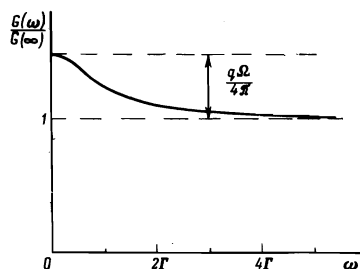


FIG. 1. Fluctuation spectrum for the intensity of spontaneous emission (semi-classical calculation).

The medium under investigation was mercury vapor. We investigated the luminescence due to the $6^3P_1 - 6^1S_0$ (2537\AA , $\Gamma^{-1} = 1.2 \times 10^{-7}$ sec) transition which was excited optically with the aid of a mercury lamp. The radiation detector was the FEU-57 photomultiplier with a quantum yield at 2537\AA amounting to $q = 5\%$ (this was the nominal figure, confirmed by direct measurement). The luminescence of the vapor was excited in a container which was designed to ensure maximum light collection onto the photomultiplier photocathode. The container was made of quartz and had an aluminized spherical outer part. The saturated vapor pressure in the container corresponded to the temperature of the cooling system. The cooling system was filled with a mixture of the solid and liquid phases of paraffins or mercury, which enabled us to hold the container for long periods of time at the melting points of nonane (-53°C), decane (-29°C), undecane (-26°C), and mercury (-39°C). At all these temperature points the mercury vapor pressure was low enough to enable us to neglect the trapping of resonance radiation and the associated theoretical complications connected with the prediction of the noise spectrum. The source of the exciting radiation was a mercury lamp, using a high-frequency discharge in a controlled mercury-vapor density. The light from this lamp was collimated into a narrow beam which was received by the entrance window of the container. The signal from the cathode was fed into an amplifier with two isolated passbands of $\Delta f_l = \omega_p/2\pi = \Delta f_h = 100$ kHz around the frequencies of 400 kHz and 4 MHz. The low- and high-frequency signals were separated by filters at the amplifier output. They were then detected and fed into a bridge comparison circuit.

The spectrum shown in Fig. 1 represents the fluctuation spectrum of the photocurrent from an ideal photoelectric detector. We used a special calibration procedure to eliminate any nonuniformity in the frequency characteristic of the photomultiplier itself and possible differences between the integrated gain in the two spectral intervals. The photomultiplier was alternately exposed to the luminescence and to equal intensity direct radiation from the mercury lamp. To achieve this, a small aperture covered by a diffuser was made in the aluminized dome of the reflector, opposite the photomultiplier window, and the radiation from the lamp was projected onto this aperture. Since the light from the lamp was collected from a small solid angle, its fluctuation spectrum was not expected to contain the low-frequency excess noise. Therefore, by balancing the bridge with the photomultiplier exposed to the radiation from the lamp, and then recording the luminescence, it was possible to estimate the required signal from the off-balance voltage. To eliminate the effect of the slow zero drift, the radiation from the mercury lamp was periodically switched from luminescence excitation to direct illumination of the photomultiplier with the aid of a disk modulator (Fig. 2). This enabled us to use synchronous detection of the voltage across the bridge diagonal, which was monitored by a strip chart recorder. When the modulator was employed, steps were taken to ensure that the radiation intensity in the two optical channels was the same (to within 0.5%), which guaranteed the absence of parasitic synchronous signals because of the highly sensitive balancing of the bridge.

We shall not pause to describe in detail the balancing and calibration procedures, and will merely summarize the data on the sensitivity of the system. The time of a

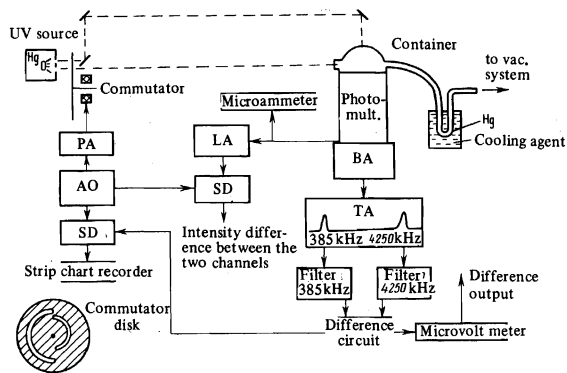


FIG. 2. Block diagram of the apparatus: PA—power amplifier, AO—audiofrequency oscillator, SD—synchronous detector, LA—low-frequency amplifier, PM—photomultiplier, BA—broadband amplifier, TA—tuned amplifier.

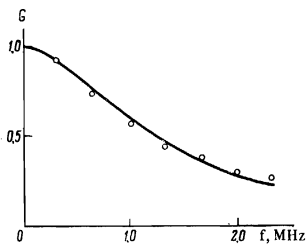


FIG. 3. Fluctuation spectrum for spontaneous emission of mercury ($\lambda = 253.7$ nm) under electron-impact excitation. Points—experimental, solid line—theoretical for $\Gamma^{-1} = 1.3 \times 10^{-7}$ sec.

single measurement was limited by the temperature stability of the cooling system and amounted to 10–15 min. With this averaging period, the sensitivity of the system was sufficient to detect systematic deviations of 0.02% in the level of mean square fluctuations, which is close to the theoretical limit.^[14]

For the final verification of the nature of the expected signals we planned an additional control operation in which we recorded luminescence which was highly quenched by a molecular gas (in our case, hydrogen). This quenching gas reduced the luminescence intensity by a substantial factor (this was compensated by an increase in the excitation intensity) and increased the damping constant Γ by the same factor.^[15] In this way, we could vary the life-time of the excited atoms, and hence the expected shape of the spectrum, without varying the other experimental conditions.

The temperature of the mercury vapor was chosen so that the luminescence intensity exceeded parasitic scattered radiation by a factor of 3–10 in all cases (this was checked by freezing out the mercury vapor). The experiments were performed in a broad range of luminescence intensities, namely, between 3×10^8 and 1.6×10^{10} photoelectrons collected per second from the photomultiplier cathode.

All the experiments gave a negative result. We did not record any systematic change (to within the limits of sensitivity, i.e., 0.02%) in the noise power difference between the chosen spectral intervals on switching from luminescence to scattered radiation. The figure of 0.02% must be compared with the theoretical estimate of the signal. Let us suppose that the collection factor $\Omega/4\pi$ was 0.5, which is an underestimate. We must also allow for the reduction in the theoretical value of the signal due to the excess noise associated with multiplication in the photomultiplier. The experimental upper limit for the noise factor of the photomultiplier was found to be 3.

Finally, since the frequency intervals $\Delta\omega_l$ and $\Delta\omega_h$ were separated by a finite distance, the theoretical noise difference between them should be 0.8 of the maximum value. If the quantum efficiency of the photomultiplier is 5%, this gives a minimum of 0.6% of the shot noise power, which exceeds by a factor of at least 30 the sensitivity within the limits of which the signal was not detected.

4. DISCUSSION

We have thus found that the semiclassical description of spontaneous emission is not consistent with experimental results which can, however, be described in terms of quantum mechanical ideas. To elucidate this statement, Appendix 2 gives a phenomenological calculation of the fluctuation spectrum for a model of the source based on the quantum mechanical description of the emission process. According to this model, there is a finite probability of detecting the photon as soon as the atom is excited, and this probability decays exponentially with time. Using this model, and neglecting interference between different atoms,^[3] we can see that the flux of recorded photons has the same behavior (with random delay, on the average amounting to Γ^{-1}) as the distribution of the excitation events in time. If the latter are uncorrelated with one another, which was the case in our experiment, then the photons originating from them cannot be correlated either. The expression for the spectrum of fluctuations in the photodetector output can in this model be written in the general form^[4]

$$G_v(\omega) = |\bar{u}(\omega)|^2 q \langle F \rangle \left[1 + q \frac{\Omega a(\omega)}{4\pi} \frac{G_J(\omega)}{\langle J \rangle} \right], \quad (9)$$

where $a(\omega)$ is determined directly by the probability of decay: $a(\omega) = |p(\omega)|^2$; $G_J(\omega)$ and $\langle J \rangle$ are, respectively, the intensity fluctuation spectrum and mean excitation intensity $J(t)$.^[5] It is clear from (9) that the spectrum of the elementary process now appears only in the product in which the spectrum of fluctuations in the excitation intensity is the other factor. If the pump is constant in time [$G_J(\omega) = 0$], the output-signal spectrum contains only the white component of shot noise.

To complete the picture, these conclusions were verified in an additional experiment with fluctuating excitation. The mercury vapor was excited by an electron beam whose intensity was modulated in accordance with a random law using a noise generator with a uniform spectrum between 0 and 6 MHz. The photocurrent fluctuation spectrum was recorded in this case by a standard spectrum analyzer. The spectral characteristics of the receiving channel and the photomultiplier were allowed for by recording the photocurrent fluctuation spectrum due to the photomultiplier itself with the mercury vapor replaced by hydrogen, whose luminescence in our case can be regarded as having zero inertia. Measurements showed that the shot component of the noise was a small fraction of the resultant photocurrent noise, so that the procedure used to find the required spectrum $a(\omega)$ from experimental data reduces to the division of the noise spectrum for the mercury luminescence by the hydrogen noise spectrum, as shown in (9). The result is given in Fig. 3, where the solid line indicates the theoretical curve.

We note, in conclusion, that the latter experiment is a demonstration of a new method of investigating the lifetimes of excited states of atoms, which is a linear variant of the method developed previously in.^[16]

APPENDIX 1

The classical radiation field due to a system of atoms or, more precisely, the complex analytic signal associated with the real field observed at the point P on the photocathode at time t, can be written in the form

$$A(t, P) = \sum_{j=1}^{N(r, P)} A_j(t, P), \quad (1.1)$$

$$A_j(t, P) = (4\pi)^{-1/2} r_j^{-1} \exp[i(\omega_j t - k r_j + \varphi_j)] v(t - t_j),$$

where $A_j(t, P)$ corresponds to the field of the j-th excited atom, i.e., a spherical wave of frequency ω_j (k_j is the wave-vector modulus) and initial phase φ_j , t_j is the time at which the atom is excited, $r_j = r(P, P_j)$ is the distance between the point of observation and the radiating atom, and $v(t - t_j)$ is a function which describes the attenuation of the wave and is zero for negative values of the argument. We shall use the normalization condition

$$\int_{-\infty}^{+\infty} v(t) v^*(t) dt = 1, \quad (1.2)$$

i.e., the total energy emitted by the atom is equal to the energy of a single photon.

The random quantities ω_j , φ_j , t_j and the vector defining the position of the point P_j have independent distributions. In particular, the phase will be assumed to be uniformly distributed, i.e.,

$$\langle e^{i\varphi} \rangle = 0. \quad (1.3)$$

To begin with, let us calculate the coherence function for the field A:

$$\begin{aligned} \Gamma(t', t''; P', P'') &= \langle A(t', P') A^*(t'', P'') \rangle \\ &= \left\langle \sum_{j=1}^N \sum_{k=1}^N A_j(t', P') A_k^*(t'', P'') \right\rangle. \end{aligned} \quad (1.4)$$

After substituting (1.1) in (1.4), the averaging over the parameters of the elementary spherical waves can be carried out separately because they are independent. In particular, averaging over the phases gives

$$\langle \exp[i\varphi_j - i\varphi_k] \rangle = \delta_{jk}. \quad (1.5)$$

Therefore

$$\begin{aligned} \Gamma(t', t''; P', P'') &= \langle N(T, V) \rangle \left\langle \exp[i\omega(t' - t'')] \right\rangle \left\langle \frac{\exp[ik(r' - r'')]}{4\pi r' r''} \right\rangle_{\omega} \\ &\quad \times \langle v(t' - t) v^*(t'' - t) \rangle_{\tau}, \end{aligned} \quad (1.6)$$

where $r' = r(P', P)$, $r'' = r(P'', P)$.

In deriving (1.6) we allowed for the fact that the number of terms in the sums in (1.4) is random. The subscripts T, V, and ω represent averaging over the times of excitation, the positions of the radiating atoms, and the distribution of the optical frequencies which henceforth will be assumed to be the Doppler distribution. The mean light intensity is obtained from (1.6) by equating the coordinates:

$$\begin{aligned} \langle I(t', P') \rangle &= \Gamma(t', t'; P', P') = \langle N \rangle \langle (4\pi r^2(P', P))^{-1} \rangle_{\omega} \\ &\quad \times \langle |v(t' - t)|^2 \rangle_{\tau}. \end{aligned} \quad (1.7)$$

If the excitation probability is independent of time and $T \rightarrow \infty$, the time dependence in (1.7) will vanish when (1.2) is taken into account.⁶⁾

We now calculate the correlation function for the instantaneous intensity of the field:

$$\langle I(t', P') I(t'', P'') \rangle = \langle A(t', P') A^*(t', P') A(t'', P'') A^*(t'', P'') \rangle$$

$$= \left\langle \sum_{j, k, l}^N A_j(t', P') A_j^*(t', P') A_k(t'', P'') A_k^*(t'', P'') \right\rangle. \quad (1.8)$$

After averaging over the phases, and by virtue of the obvious result

$$\langle \exp[i(\varphi_j - \varphi_k + \varphi_l - \varphi_l)] \rangle = \delta_{ij} \delta_{kl} + \delta_{il} \delta_{jk} - \delta_{ij} \delta_{kl} \delta_{jk} \delta_{il} \quad (1.9)$$

the multiple sum in (1.8) splits into combinations of double and single sums. The single sums contain equally distributed terms and are averaged in an obvious fashion. When the double sums are averaged it must be remembered that the 'diagonal' elements (equal labels) are distributed differently from the nondiagonal elements. Omitting the relatively elementary transformations, we give the final results for the second moment of the field intensity:

$$\begin{aligned} \langle I(t', P') I(t'', P'') \rangle &= \langle I(t', P') \rangle \langle I(t'', P'') \rangle + |\Gamma(t', t''; P', P'')|^2 \\ &\quad + \langle N \rangle \left\langle \frac{|v(t' - t)|^2 |v(t'' - t)|^2}{(4\pi r' r'')^2} \right\rangle. \end{aligned} \quad (1.10)$$

In deriving this expression we used the condition

$$\langle N(N-1) \rangle = \langle N \rangle^2.$$

It is thus seen that the correlation function for the field intensity in the above model of the source consists of the two terms

$$\begin{aligned} |\Gamma(t', t''; P', P'')|^2 + \langle N \rangle \langle (4\pi r' r'')^{-2} |v(t' - t)|^2 \\ \times |v(t'' - t)|^2 \rangle, \end{aligned} \quad (1.11)$$

the first of which describes the spatial (for $t' = t''$ and $P' \neq P''$) and temporal (for $T' \neq T''$ and $P' = P''$) intensity beats (these are the Hanbury Brown-Twiss and the Rebka and Pound effects), and the second term gives the specific time beats connected with the fact that the model which we are considering differs from the Gaussian model of the field.^[11]

Further calculations connected with the substitution of (1.11) into (5) in the main text depend on the geometry of the experiment, since we have to integrate over the cathode of the photodetector and through the volume of the source. To obtain clearer results, we shall simplify the situation by supposing that the linear size of the source is small in comparison with the distance to the photosensitive surface. If the points P' and P'' lie in the plane of the detector, and are not too far from one another in comparison with the distance of the source, we can write the normalized coherence function in the form

$$\begin{aligned} \gamma(t', t''; P', P'') &= \Gamma(t', t''; P', P'') [\langle I(t', P') \rangle \langle I(t'', P'') \rangle]^{-1/2} \\ &= \langle \exp[i\omega(t' - t'')] \rangle \langle \exp[i\omega(r' - r'')/c] \rangle_{\omega} \langle v(t' - t) v^*(t'' - t) \rangle_{\tau}. \end{aligned} \quad (1.6a)$$

The assumption that the optical spectrum is relatively narrow enables us to separate the temporal and spatial coherence factors:

$$\gamma = \langle \exp[i\omega(t' - t'')] \rangle_{\omega} \langle \exp[i\omega_0(r' - r'')/c] \rangle_{\omega_0} \langle v(t' - t) v^*(t'' - t) \rangle_{\tau}. \quad (1.6b)$$

Hence the correlation function for the light-flux fluctuations on the receiver is given by

$$\begin{aligned} K_{\mathcal{F}}(t', t'') &= |\langle \exp[i\omega(t' - t'')] \rangle_{\omega}|^2 \langle v(t' - t) v^*(t'' - t) \rangle_{\tau}^2 \\ &\quad \times \int_S \int_S \langle I(t', P') \rangle \langle I(t'', P'') \rangle |\exp[i\omega_0(r' - r'')/c]|^2 ds' ds'' \\ &\quad + \langle |v(t' - t)|^2 |v(t'' - t)|^2 \rangle_{\tau} \int_S \int_S \langle I(t', P') \rangle [4\pi(r'')^2]^{-1} ds' ds''. \end{aligned} \quad (1.12)$$

Assuming that the coherence area is small, i.e.,

$$\sigma = \int |\langle \exp[i\omega_0(r' - r'')/c] \rangle_{\omega_0}|^2 ds' \ll S,$$

and neglecting edge effects, we can transform (1.12) to the form

$$K_F(t'-t'') = |\langle \exp [i\omega(t'-t'')] \rangle_\omega|^2 \times |\langle v(t'-t)v^*(t''-t) \rangle_\tau|^2 \bar{F}\sigma\bar{I} + \langle |v(t'-t)|^2 |v(t''-t)|^2 \rangle_\tau \bar{F}\Omega/4\pi, \quad (1.12a)$$

where Ω is the solid angle subtended by the photocathode at the center of the source, and \bar{I} and \bar{F} are, respectively, the mean illuminance and radiation flux in the plane of the detector.

We now obtain the fluctuation spectrum via the Fourier transformation, remembering that the Doppler broadening is usually much greater than the natural width of the radiating state:

$$G_F(\omega) = g(\omega) \bar{F}\sigma\bar{I} + a(\omega) \bar{F}\Omega/4\pi. \quad (1.13)$$

In this expression $g(\omega)$ is the Fourier transform of the "Doppler term" $|\langle \exp(i\omega t) \rangle_\omega|^2$ (practically the Doppler profile but with half-width greater by a factor of $\sqrt{2}$ than usual) and

$$a(\omega) = \left| \int_0^\infty e^{i\omega\tau} |v(\tau)|^2 d\tau \right|^2.$$

The spectrum $a(\omega)$ has a width of the order of the natural width of the radiating state and, therefore, at frequencies of the same order, the function $g(\omega)$ can be replaced by

$$g(0) = \int |\langle e^{i\omega\tau} \rangle_\omega|^2 d\tau = \tau_c,$$

which is equal to the coherence time defined in the usual way. Moreover, recalling the definition of the degeneracy parameter $\delta = \sigma\tau_c\bar{I}$, we finally obtain equation (7) of the main text.

APPENDIX 2

Let us now investigate the time statistics of fluctuations in the photon flux incident on the photodetector for a simple quasicorpuscular model of the source. In particular, we shall suppose that a particular pump excites the atoms at random instants of time after which the atoms decay independently in such a way that the decay probability depends on the time measured from the instant of excitation. Each decay is accompanied by the emission of a photon which is recorded by the photodetector with probability $q' = q\Omega/4\pi$. In this model, we ignore the interference between the different atoms, which is unimportant for our purposes [see the discussion of equation (7) in the main text].

As before, we subdivide the time scale into sufficiently small and equal intervals $\{\Delta_k t\}$ where k are integers between $-\infty$ and $+\infty$ (we are considering the entire time axis). We shall use the following notation: r_j —number of atoms excited in the interval $\Delta_j t$, n_{kj} —number of atoms excited in the interval $\Delta_j t$ and decaying in the interval $\Delta_k t$, $n_k = \sum_j n_{kj}$ —number of atoms decaying in the interval $\Delta_k t$ (this is equal to the number of emitted photons), p_α ($\alpha = \dots, -2, -1, 0, +1, +2, \dots$)—probability of decay of the atom after α intervals following excitation where p_α if $\alpha < 0$, and

$$\sum_{\alpha=-\infty}^{\infty} p_\alpha = \sum_{\alpha=0}^{\infty} p_\alpha = 1.$$

Since the atoms decay independently of one another, the numbers n_{kj} have the distribution

$$P(\dots, n_{kj}, \dots; r_j) = r_j! \prod_{k=-\infty}^{\infty} \frac{(p_{k-j})^{n_{kj}}}{n_{kj}!}, \quad (2.1)$$

where $\sum_k n_{kj} = r_j$ and we suppose that $0^0 = 1$.

Since we are interested in the distribution of the numbers n_k , which are the sum of independent random num-

bers n_{kj} , it is natural to use the generating-function formalism. The generating-moment function for the distribution (2.1) is

$$\Phi_{r_j}^{(1)}(\dots, z_k, \dots) = M_{r_j, z_k^{n_k}} = \left(\sum_{k=-\infty}^{\infty} p_{kj} z_k \right)^{r_j} = Z_j^{r_j}, \quad (2.2)$$

where the mathematical expectation is evaluated for fixed r_j . Since the n_{kj} are independent, the generating function for the numbers n_k is

$$\Phi_{r_j}(\dots, z_k, \dots) = M_{r_j, z_k^{n_k}} = \Pi Z_j^{r_j}. \quad (2.3)$$

We must now define the statistics for the numbers r_j which govern the rate of creation of excited particles. We shall assume the Poisson statistics [which satisfies the more general condition given by (4)], i.e.,

$$P(\dots, r_j, \dots) = \Pi \frac{e^{-\mu_j} \mu_j^{r_j}}{r_j!}, \quad (2.4)$$

where $\mu_j = J(t_j)\Delta_j t$ is the mean number of excited atoms in the interval $\Delta_j t$ and $J(t)$ is the intensity of the excitation process. The generating-moment function for the numbers n_k is obtained by averaging over the distribution given by (2.4):

$$\begin{aligned} \Phi(\dots, z_k, \dots) &= M_{z_k^{n_k}} = M \Phi_{r_j} = \exp \left[\sum_j \mu_j (Z_j - 1) \right] \\ &= \exp \left(\sum_k z_k \nu_k - \sum_j \mu_j \right), \\ \nu_k &= \sum_j p_{k-j} \mu_j. \end{aligned} \quad (2.5)$$

Using the normalization $\sum_j \mu_j = \sum_k \nu_k$, we obtain the generating function in terms of only the parameters $\{\nu_k\}$:

$$\Phi(\dots, z_k, \dots) = \exp \left(\sum_k z_k \nu_k - \sum_k \nu_k \right). \quad (2.6)$$

It is readily verified that this function corresponds to the Poisson statistics (2.4) with modified mean occupation numbers ν_k . The intensity of the decay process is related to the intensity of the excitation process by the formula

$$I(t) = \int_{-\infty}^t J(t') p(t-t') dt, \quad (2.7)$$

which is obtained from (5) by passing to the limit of infinitesimal subdivision $\{\Delta_k t\}$ where $p(\tau)d\tau$ is the probability of decay in the interval $(\tau, \tau + d\tau)$ after excitation at time $\tau = 0$.⁷⁾ Therefore, the random delay of the decay event relative to the excitation event preserves the Poisson statistics and the intensity is "smoothed" by the delay distribution function $p(\tau)$.

The photoelectron statistics is connected with the photon statistics by the binomial distribution: the probability of detecting m photoelectrons during the emission of n photons is

$$B(m; n) = \frac{n!}{m!(n-m)!} (q')^m (1-q')^{n-m}, \quad m \leq n, \quad (2.8)$$

$$B(m; n) = 0 \quad m > n$$

It is known that, under the transformation defined by (2.8), the Poisson process remains a Poisson process, but the intensity is multiplied by q' , i.e., the effective quantum yield of the photodetector. It follows that the discussion used in the derivation of (6) is valid with the obvious modifications which lead to (9).

¹⁾An example of this kind of qualitative discrepancy is provided by the recent paper by Clauser. [10]

²⁾In particular, the use of the discrete detector to observe the decay of a single atom cannot resolve the dilemma formulated in the introduction.

³See the discussion of Eq. (7), $q\delta \ll 1$.

⁴See Appendix 2.

⁵We emphasize that "excitation intensity" is to be understood as the intensity of the corresponding Poisson process for the distribution of the excitation events in time.

⁶Equations (1.6) and (1.7) are valid for any interval T and an arbitrary distribution of the excitation times within this interval. In the limiting transition $T \rightarrow \infty$, it is assumed that the excitation is time-independent and the rate of excitation is finite, i.e.,

$$\lim_{T \rightarrow \infty} \frac{\langle N \rangle}{T} > 0.$$

⁷ $p(\tau)$ is also the distribution density for the lifetime of the atom in the excited state.

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130