ON THE POSSIBILITY OF DETERMINING THE SPECTRAL CONTOUR OF AN ELEMENTARY RADIATIVE PROCESS UNDER CONDITIONS OF EXCESSIVE DOPPLER BROADENING

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It is shown by elementary means that the noise spectrum of a photodetector illuminated by spontaneous radiation from an atomic system contains information on the fine structure and proper width of the radiative state despite excessive Doppler broadening. The information can be successfully derived only under the condition of a sufficiently large ratio between the populations of the initial and final combining states and an appreciable optical width of the radiating medium layer. A rough calculation of the signal to noise ratio is performed for the noise analysis operation.

It was shown earlier[1,2] that an analysis of the noise spectrum of radiation subjected beforehand to coherent amplification makes it possible to reveal in emission spectra fine details that are completely masked (in the usual spectroscopic techniques) by the Doppler broadening. Information concerning the spectral structure of the individual radiative processes is retained in the total radiation field of an ensemble of atoms, but to extract this information it is necessary to perform the operation of coherent amplification of light, the meaning of which reduces to a similar increase of the field energy while retaining its correlation properties. The amplification brings the field closer to the classical one and makes it possible to analyze this field, without loss of information, by using photoelectric receivers. However, the need for amplification greatly limits the methodological value of such an analysis. We discuss in this article the possibility of realizing a spectral noise analysis without using a coherent amplifier.

Let us consider a two-level system with nonzero populations of the upper (n_1) and lower (n_2) states. No matter what the population ratio, for a photon with resonant frequency there is a probability of absorption and a probability of stimulated emission with production of a photon pair[1)]. Therefore light passing through a medium with non-empty upper level must inevitably be "degenerated": groups of identical photons will appear in it. The presence of such groups is a sufficient condition for effecting the analysis of the intra-Doppler structure of the primary radiation[1]. At the same time, the condition for the population of the excited state is incomparably less stringent than the population inversion condition formulated in[1,2].

Bearing in mind an approach to probable experimental conditions, we consider the following one-dimensional problem. We have a medium with specified ratio of level populations, in which intrinsic spontaneous emission propagates. It is required to establish the degree of "degeneracy" (in the sense indicated above) of the radiation in a given section of the medium. The radiation is registered along the x axis, and the medium extends from 0 towards positive values of x. It is easy to see that a change in the number N_1 of the single photons, in the number N_2 of photon pairs, the number N_3 of groups of three, etc., is given by the following system of differential equations:

$$dN_1/dx = -(k_- + k_+)N_1 + 2N_2k_- + \alpha,$$
$$dN_2/dx = -2(k_- + k_+)N_2 + 3N_3k_- + k_+N_1,$$
$$\cdots\cdots\cdots$$
$$dN_j/dx = -j(k_- + k_+)N_j + (j+1)N_{j+1}k_- + (j-1)N_{j-1}k_+. \quad (1)$$

here k_+ and k_- are the separated amplification and absorption coefficients, which are proportional to n_1 and n_2. It is assumed that k_+ and k_- do not depend on x, i.e., the saturation is assumed to be negligible. The summary absorption coefficient k is equal to the difference of these coefficients: $k = k_- - k_+$.

With the exception of the first, all the equations of the system are constructed in the same manner: the number N_j of the groups containing j photons each decreases in the section dx as the result of absorption in the medium (with transition to the lower group $j - 1$), and as the result of stimulated emission (with transition to the group $j + 1$). At the same time, the number of groups N_j increases because of the influx of neighboring groups from below and from above, as the result of stimulated emission and absorption. An exception is the first equation, in which the constant α takes into account the spontaneous emission of single photons.

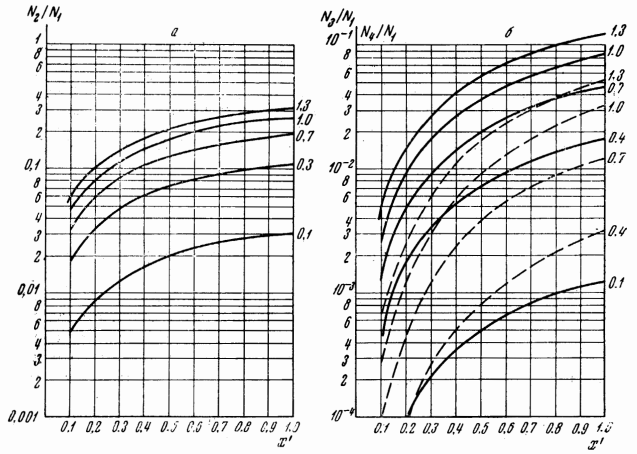
The correctness of the system of equations (1) can be verified by summing all the equations with factors corresponding to the numbers of the groups, which leads to the usual Bouguer equation in differential form:

$$\frac{d}{dx}\sum_{j=1}^{\infty} jN_j = -k\sum_{j=1}^{\infty} jN_j + \alpha,$$

since $\sum_{j=1}^{\infty} jN_j = N$ is the total number photons in the given section.

Obviously, the system of coupled equations (1) can be terminated and its solution will be valid for not too thick layers of the medium. A criterion for the validity may be satisfaction of the Bouguer law for the partial sum of photons $\sum_{j=1}^{l} jN_j$. An equivalent condition is the

1)It must be emphasized that here and throughout the word "photon" means a single-photon state of the field [3], having no strictly defined energy and described by a wave packet. Such a photon arises in the elementary act of emission of the atom.



Dependence of the relative number of photon groups on the length of the radiating layer and on the ration of the populations of the combining levels: a—doublets, b—triplets and quartets (dashed).

requirement $N_l \ll N_{l-1}$, where l is the number of the highest group that remains under consideration.

The figure shows a plot of the solution for three photon groups, obtained with the aid of a computer[2)]. The abscissa represents the reduced optical density of the layer $x' = k_-x$. The ordinate represents the ratio of the number of groups of photons with given number to the number of single photons. On the right of each curve is marked the ratio $\beta = k_+/k_- = n_1/n_2$ of the level populations.

The system (1) makes it possible to obtain asymptotic solutions at large densities x in the region $\beta < 1$. In this region the medium as a whole is absorbing, and therefore, at sufficiently large distances $(kx > 1)$ the light flux becomes stationary and the system (1) turns into an infinite system of algebraic equations. This system admits of the exact solution

$$N_j / N_1 = \beta^{j-1} / j. \quad (2)$$

From this solution we get, in particular, the curious deduction that when $\beta \to 1$ the numbers of the photons gathered into groups cease to depend on the number of the group.

It is now necessary to connect the obtained results on the grouping of the photons with the observed quantity—the excess noise in the spectrum of the receiver photocurrent. The excess noise connected with the interference between the harmonics of the elementary radiative process should not be too small relative to the background due principally to the shot noise.

Let the intensity of the elementary process of emission follow a time law f(t). This means that the wave packet accompanying the photon emitted by the atom is assigned this law. Unlike the central optical emission frequency, which varies randomly from atom to atom within the Doppler line width, the law f(t) is of the same type for all atoms, since it is determined mainly by their individuality and by the external field in which they are situated. Assume, further, that after the initial photon passes through the medium it serves as a nucleus of a group of j photons, and then the intensity of the group is proportional, as before, to f(t). The arrival of the photon group to the photocathode of the receiver leads to emission of p correlated photoelectrons $(p \le j)$. The average power spectrum $\langle g^p(\omega) \rangle$ of this group of electrons is given by[1]

$$\langle g^p(\omega) \rangle = p\varphi(\omega) + \varphi(\omega)(p^2 - p)\langle g_0(\omega) \rangle,$$

where $\varphi(\omega)$ is the frequency characteristic of the receiver and $\langle g_0(\omega) \rangle$ is the average power spectrum of the initial process f(t). The quantity $\langle g^p(\omega) \rangle$ is the contribution made to the photocurrent spectrum by the groups of p photoelectrons each. Since the arrival of each group occurs independently, the total photocurrent spectrum $G(\omega)$ is the sum of all the partial spectra:

$$G(\omega) = \sum_{p=1}^{\infty} \eta_p \langle g^p(\omega) \rangle. \quad (3)$$

We have introduced here the symbol η_p—the number of groups of p photoelectrons each, emitted by the cathode per unit time. The numbers η_p are connected with the numbers N_j of the photon groups via the quantum yield q of the receiver. The probability $P_j(p)$ for the emission of p photoelectrons under the influence of a group of j photons is given by the binomial distribution

$$P_j(p) = \frac{j!}{(j-p)!\,p!} q^p (1-q)^{j-p}.$$

Thus

$$\eta_p = \sum_{j=p}^{\infty} P_j(p) N_j.$$

Going over to estimates, we shall consider the unfavorable (but apparently realistic) situation characterized by a large excess of the population of the lower state over the upper one $(\beta \ll 1)$. Under these conditions, as seen from the figure and from the stationary solution (2), we can consider only single photons and pair groups (i.e., j = 1, 2). Accordingly, formula (3) for the total noise simplifies to

$$G(\omega) = qN_1\varphi(\omega) + 2q^2N_2\varphi(\omega)\langle g_0(\omega) \rangle. \quad (4)$$

The information of interest to us, concerning the individual spectrum $\langle g_0(\omega) \rangle$, is contained in the second term of (4), whereas the first term corresponds to the shot noise. It follows from (4) that the ratio of the useful component of the noise to the useless shot noise is given by the coefficient

$$\varkappa = 2q\langle g_0(\omega) \rangle N_2 / N_1.$$

The normalized spectral power $\langle g_0(\omega) \rangle$ does not exceed unity. Since it is assumed that $N_1 \gg N_2$ and that $q < 1$ always, κ is always much smaller than unity.

Before we proceed to consider a concrete example, in which the final signal to noise ratio will be obtained with allowance for the properties of the object and the apparatus, we must make a general remark. The developed simple approach to the problem, strictly speaking, is valid only at such a low radiation intensity when the wave packets of the emission of different atoms do not overlap in either space or in time. Otherwise it is necessary to take into account the interference between them. However, it is well known[4] that such an inter-

[2)]The computer calculations were made by A. V. Burlakov and I. N. Taganov, to whom the author is deeply grateful for collaboration.

ference leads to the appearance of excess noise with a very broad spectrum (on the order of double the width of the Doppler line). Consequently, such an additional noise does not interfere with the observation of relatively narrow spectral maxima $\langle g_0(\omega)\rangle$ of the individual spectrum. The presence of an additional Doppler spectrum likewise does not change the obtained quantitative results, since the power of the Doppler noise is always much smaller than the power of the shot noise, relative to which the estimate was made. (The ratio of the power of the Doppler noise to the shot noise is numerically equal to the degeneracy parameter of the field[5], which in the visible region, for non-laser sources, does not exceed 10^{-3}.)

Let us consider as an example the problem of separating the individual spectrum. Let the emission spectrum of each atom consist of two close lines of equal intensity, which are the result, for example, of magnetic splitting. The power spectrum of the emission in the elementary act, averaged over the ensemble of emitters, is given by[1]

$$\langle g_0(\omega)\rangle = \Gamma^2(\Gamma^2+\omega^2)^{-1} + 2^{-2}\Gamma^2\,[\Gamma^2+(\omega-\omega_{12})^2]^{-1} + 2^{-2}\Gamma^2[\Gamma^2+(\omega+\omega_{12})]^{-1}.$$

Here Γ is the natural width of both initial emitting states. The spectrum $\langle g_0(\omega)\rangle$ has two maxima, one in the region $\omega = 0$, connected with the interference of the harmonics within the limits of the width of each line, and the other in the region $\omega = \omega_{12} = \omega_1 - \omega_2$ (ω_1 and ω_2 are the optical frequencies of the two lines), which is smaller by a factor of 4 than the quantity resulting from the beats of the harmonics of one of the lines with the harmonics of the other. Since the position of the second maximum can be varied over the spectrum by varying the splitting ω_{12}, the second maximum is methodologically easier to observe, even though it is smaller.

In the vicinity of the beat frequency ω_{12}, the ratio of the excess spectral power to the shot-noice background is equal to $\kappa = qN_2/2N_1$. The excess noise can be measured by a standard technique. A spectral slit of width Δf is cut out from the noise spectrum, and governs the resolution of the system. The noise passed by the slit is detected with a certain time constant Δt, and this signal at the detector output is compared with the readings of the detector in the absence of the excess noise. It is known[6] that the mean-squared ratio of the obtained signal to the fluctuations of the background is determined under these conditions by the quantity $S = \kappa\sqrt{\Delta f\,\Delta t}$.

Let us make a numerical estimate. We take a system with a parameter $\beta = 0.1$ and a sufficiently large optical density, so as to be able to use the stationary solution $N_2/N_1 = \beta/2$. Assume, further, that the quantum yield of the receiver is 0.15. We specify a resolution of 10^5 Hz (3.3×10^{-6} cm^{-1}), which is adequate for practically all spectroscopic needs. Then at a measurement time $\Delta t = 100$ sec we obtain for the final signal to noise ratio a value somewhat larger than 10.

Thus, under conditions which are not particularly extraordinary, the foregoing procedure makes it possible to observe a spectral structure which is not accessible to direct spectroscopy methods, and to obtain information concerning the natural width of the term. The procedure does not require a special preparation of the excited state of the radiating atoms, as is the case when such methods as double resonance, intersection of levels, and others are used. A characteristic feature of this method is the independence of the relative magnitude of the signal on the brightness of the source, if the main noise is of the shot type. The latter condition, as a rule, can be readily satisfied if a photoelectronic multiplier is used.

In conclusion it should be noted that the described analysis principle is very simple methodologically and is based on the use of standard radio apparatus. The extent to which this method can be applied depends on the extent to which it is possible to realize, for a wide group of objects, a medium with an appreciable value of the parameter β and with sufficient optical density. This question calls for a separate study. We can only indicate that the population ratio considered in the example is established at a system temperature 2—3 eV (visible band). At the same time it is known with increasing free-electron density the distribution of the populations of the electron levels in a gas discharge approaches the thermal distribution corresponding to the temperature of the free electrons. On the other hand, the electron temperature usually amounts to several electron volts, and this supports our optimism in the estimate of the possibilities of the noise spectral analysis.

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