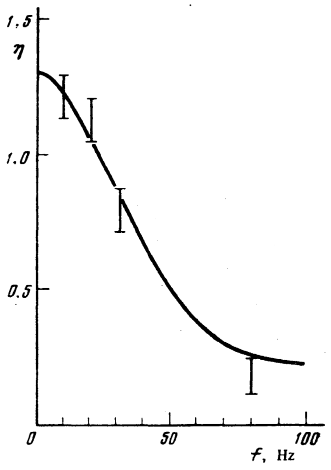

FIG. 3. Frequency dependence of absorption noise level.

the control cell (b), and the noise level when the detector was exposed to the hot-filament lamp (c). The photocurrent flowing through the receiver was the same in all cases. The ratio η was found by dividing the difference between the squares of the mean signal levels for (a) and (b) by the squares of the mean signal level for (c). Figure 3 shows the frequency dependence of η.

4. The experimental data are compared in Fig. 3 with the calculated curve given by the Lorentz formula $I^2_\omega = I^2_\omega(0)/(\omega^2\tau_c^2+1)$ corresponding to a very approximate description of diffusion by the single constant $\tau_c = 0.025$ sec. Precise agreement with experiment was not expected but, in fact, the agreement is quite reasonable as a first approximation.

The experiment has thus confirmed the presence of appreciable fluctuation in the absorption by atomic vapor. The absolute magnitude of this noise and its spectral distribution are in qualitative agreement with expectations. Noise of this origin may be important in experiments on the optical pumping of atoms with buffer gases because the pumping time T is then usually chosen to be comparable with the phase relaxation time T_2 which, as a rule, is much smaller than the diffusion time τ_c. This noise is concentrated in the neighborhood of a few tens of hertz, and this must be taken into account in choosing the detection bandwidth. Shot noise may also appear in laser systems for the stabilization of frequency, using saturated absorption. The small cross section of the absorbing atoms is, in this case, compensated by high illumination intensity.

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Statistics of thermal fluctuations of optical absorption of gases

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A theory of thermal fluctuations of the optical absorption of a gas is presented. It is shown that the light passing through a layer of absorbing gas contains, besides the shot noise, additional noise connected principally with fluctuations in the number of absorbing atoms in the volume occupied by the light beam. Expressions are obtained for the spectrum and power of the excess noise.

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Aleksandrov and Mamyrin[1] have performed an experiment aimed at observing the fluctuations of the intensity of a light beam passing through a layer of an absorbing gas. It was found that these fluctuations contain, besides the photon component proper, a contribution connected with the absorption fluctuations due to the thermal motion of the gas atoms. The power spectrum of these additional fluctuations will be calculated below, and quantitative estimates will be obtained of their observability against a background of the shot fluctuations of the radiation.

Let a photon beam with initial intensity I_0 photons per second, uniformly distributed over a cross section S, traverse a path l in a cell with the absorbing gas. Neglecting the travel time l/c, we relate the probability τ of free flight of the photon with a definite instant of time t. Assuming the initial beam to have a Poisson distribution and that the photons are absorbed independently, we obtain at the exit from the cell a bistochastic Poisson flux with random intensity $I(t) = I_0\tau(t)$. As shown in[2], the noise spectrum of a photodetector illuminated by such a photon flux is

$$G(\omega)=|\tilde{u}(\omega)|^2[qI_0\overline{\tau(t)}+q^2I_0^2G_\tau(\omega)], \tag{1}$$

where $\tilde{u}(\omega)$ is the frequency characteristic of the photodetector, q is its quantum yield, and $\overline{\tau(t)}$ and $G_\tau(\omega)$ are the mean value and the power spectrum of the modulating process $\tau(t)$.

To calculate the required first two moments of the process $\tau(t)$, it is necessary to assume a certain model that connects the probability τ of the photon free path with the distribution of the absorbing atoms and their number in the transilluminated volume $V=Sl$ of the cell. We assume the simplest shadow model, assuming the atoms to be opaque particles with an absorption cross section σ that is randomly distributed in space and varies its position as a result of independent thermal motion. An obvious shortcoming of this model is that it does not agree with the wave picture of light propagation, but it can be easily used to obtain the principal part (see below) of the observed effect.

Within the framework of the assumed model we have

$$\tau=\int_S \varphi(Q)\,dS_Q/S, \tag{2}$$

where Q is a point in the plane of the entrance face of the cell; $\varphi(Q)=0$ if the shadow projection of all the absorbing particle in the volume on the plane of the end face covers the point Q, and $\varphi(Q)=1$ in the opposite case.

Assuming the atoms to have a Poisson spatial distribution with an average concentration C [cm^{-3}] we easily obtain

$$E\varphi(Q)=P(n(v_Q)=0)=e^{-Cv_Q}=e^{-M\sigma/S}, \tag{3}$$

where E and P are respectively the symbols for the mathematical expectation value and the probability, $n(v_Q)$ is the number of particles in the cylindrical volume $v_Q=\sigma l$ whose axis passes through the point Q, and $M=CV$ is the average number of particles in the volume, i.e.,

$$\bar{\tau}(t)=E\tau=e^{-k}, \tag{4}$$

where $k=M\sigma/S$ denotes the optical density of the cell.

Proceeding to the calculation of the second moments of the process $\tau(t)$, we note that the absorption fluctuations described by the assumed model are determined by two different factors: 1) the fluctuations of the total number of absorbing particles $m=n(V)$ in the volume V, and 2) the fluctuations of the mutual placement (configuration) of the particles at a fixed value of m.

To estimate the relative role of the indicated components, we represent the total variance of the quantity τ in the form of two terms

$$D(\tau)=E\tau^2-(E\tau)^2=ED_m(\tau)+D(\tau_m), \tag{5}$$

where $\tau_m=E_m\tau$ is the conditional mean value of τ at a fixed number of particles $m=n(V)$, $D_m(\tau)=E_m\tau^2-\tau_m^2$ is the conditional variance, and $D(\tau_m)=E\tau_m^2-(E\tau_m)^2$ is the variance of the conditional mean value. The first term in (5) describes the contribution of the configuration fluctuations, whereas the second represents the contribution of the fluctuations of the number of particles. Simple calculations based on the representation (2) and on Poisson statistics of the occupation numbers yield

$$\tau_m=(1-\sigma/S)^m,\quad D(\tau_m)=e^{-2k}(e^{k\sigma/S}-1). \tag{6}$$

Using the obvious relation

$$E\varphi(Q)\varphi(Q')=P(n(v_Q\cup v_{Q'})=0),$$

where $v_Q\cup v_{Q'}$ is the union of the cylindrical volumes σl centered respectively at the points Q and Q', we can obtain the representation of the configuration term in (5):

$$ED_m(\tau)=e^{-2k}\frac{\sigma}{S}\int_S\left(e^{kS(Q)/\sigma}-e^{k\sigma/S}\right)\frac{dS_Q}{\sigma}. \tag{7}$$

Here $S(Q)$ is the area of the overlap of the shadow projections of two particles, one of which is fixed on the beam axis and the other is projected into the point Q. Comparison of (6) and (7) shows that both components of the total variance are of the same order in terms of the parameter σ/S. Moreover, expanding the exponential under the integral sign and estimating it term by term (after integration), we obtain an estimate of the relative value of the configuration variance

$$\frac{ED_m(\tau)}{D(\tau_m)}\leqslant\frac{e^k-1-k}{k}=\frac{k}{2}+\frac{k^2}{6}+\frac{k^3}{24}+\ldots \tag{8}$$

In the considered experiment we have $k\approx1$, i.e., both components of the variance are approximately equal in magnitude.

Proceeding now to an examination of the dynamics of the process $\tau(t)$, we note that the change of the particle configuration in the volume V and the change of their total number $m(t)$ occur at substantially different rates. The characteristic time of variation of the configuration of the gas particles is the time required for the particle to be displaced by an amount equal to its diameter (in the "shadow" model) or by a distance on the order of the wavelength (with allowance for wave effects). In either case, this time is quite short (10^{-10}–10^{-8} sec in the discussed experiment), and that fraction of the total variance of the absorption fluctuations which is connected with this factor will be distributed in the power spectrum of the $\tau(t)$ process over a wide frequency interval. On the other hand, the fluctuations of the number of particles are determined by the Brownian motion of the absorbing atoms and can be made slow enough (for example, of the order of 10^{-2} sec) by adding a buffer gas. Then, bearing in mind the observation of the fluctuations at relative low frequencies, we can neglect completely the configuration component of the noise.[1)]

By virtue of the foregoing arguments, we shall henceforth estimate the spectral properties of the process $\tau(t)$ defined in the following manner:

$$\tau_m(t)=\tau_{m(t)}=(1-\sigma/S)^{m(t)}.$$

Assuming (see below) that the number of particles in the volume V is subject to Poisson statistics at an arbitrary instant of time, we obtain, by using the technique of generating functions for the moments, a relation between the correlation functions $K_\tau(t)$ and $K_m(t)$ of the process $\tau(t)$ and $m(t)$:

$$K_\tau(t)=e^{-2k}\left[\exp\left\{\left(\frac{\sigma}{S}\right)^2 K_m(t)\right\}-1\right]=e^{-2k}\left(\frac{\sigma}{S}\right)^2 K_m(t),\quad \frac{\sigma}{S}\ll 1. \tag{9}$$

To analyze the statistics of the process $m(t)$ we assume that the volume V transilluminated by the photon beam constitutes an (infinitesimally) small part of a certain reservoir with gas. Assume that at the initial instant of time the gas particles in this reservoir have a Poisson spatial distribution with an average volume concentration $C_0(r)$ [cm^{-3}], that depend perhaps on the coordinates r. The Brownian motion of an individual particle is described by a Markov random process with a transition probability density $f(r, t; r_0, t_0)$,[3] which determines the density of the probability distribution of the coordinate r of the diffusing particle at the instant of time t, under the condition that at the preceding instant of time $t_0<t$ the particle was at the point r_0. Using a generating-function technique analogous to that employed in[2], we can show that in the case of independent diffusion of the atoms they retain a Poisson spatial distribution with a volume concentration

$$C_t(r)=\int f(r,t;r_0,t_0)C(r_0)\,dv_0.$$

By the same method, taking into account the explicit form of the transient density for three-dimensional diffusion,[3] we usually obtain the following expression for the correlation function of the process $m(t)$:

$$K_m(t)=\frac{C}{(2\pi Dt)^{3/2}}\int\limits_V\int\limits_V \exp\left\{-\frac{(x-x')^2+(y-y')^2+(z-z')^2}{2Dt}\right\}dx\,dy\,dz\,dx'\,dy'\,dz', \tag{10}$$

where D is the diffusion coefficient and (x, y, z) or (x', y', z') are the Cartesian coordinates describing the volume V.

Thus, the exact form of the function $K_m(t)$ can be obtained by solving the diffusion problem for the region V. We confine ourselves to a qualitative treatment, our aim being to obtain the time scale of the function $K_m(t)$, inasmuch as the details of the behavior of this function depend on the structure of the light beam in the gas cell and could not be followed experimentally in practice. We assume that the beam is in the form of a circular cylinder of radius R and axis directed along the z axis. Then, integrating with respect to z and z' and changing over to dimensionless variables, we easily obtain

$$K_m(t)=MF(\Gamma t), \tag{11}$$

where $M=C\pi R^2 l$ is the average number of particles in the volume V, $\Gamma=D/R^2$ [sec^{-1}] plays the role of the time constant, and the function $F(t')$ of the dimensionless argument t' is determined by the double integral

$$F(t')=\frac{1}{2\pi^2 t'}\int\limits_{S_1}\int\limits_{S_1}\exp\left\{-\frac{(x-x')^2+(y-y')^2}{2t'}\right\}dx\,dy\,dx'\,dy'$$

over the area of the unit circle S_1, $F(0)=1$.

Substituting (4) and (11) in (1), we obtain the final expression for the observed spectrum of the fluctuations

$$G(\omega)=|\tilde{u}(\omega)|^2 qI_0e^{-k}\left[1+qI_0e^{-k}k\frac{\sigma}{S}\frac{1}{\Gamma}\tilde{F}\left(\frac{\omega}{\Gamma}\right)\right], \tag{12}$$

where $\tilde{F}(\omega)$ denotes the Fourier transform of the function $F(t')$. It is easily seen that $\tilde{F}(0)$ is a numerical constant and the condition for being able to observe the absorption fluctuations against the background of the shot fluctuations is given by the quantity

$$\eta=qI_0e^{-k}k\frac{\sigma}{S}\frac{1}{\Gamma},$$

which should be (taking $\tilde{F}(0)$ into account) not too small in comparison with unity. The result agrees with the estimate given in[1].

1)The foregoing calculations were made in the "shadow" approximation, i.e., no account was taken of the wave effects of light propagation in a medium with discrete absorbers. One can expect the wave effects to lead only to a smoothing of the configuration noise, i.e., to further decrease of their spectral power, whereas fluctuations of the number of the absorbers manifest themselves equally both in the wave and in the corpuscular picture of the light field.

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