# Thermal fluctuations in the optical absorption of gases

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It is shown experimentally that the intensity of a beam of light passing through an absorbing gas is modulated in accordance with a random law, owing to thermal fluctuations in the number of absorbing atoms. The fluctuation spectrum is localized in the region 0 to $1/2\pi\tau$ Hz, where $\tau$ is the characteristic time for the diffusion of an atom through the beam cross section. Theoretical estimates of the absorption fluctuation power are in agreement with experimental data.

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1. In our previous publication[1-4] we considered the possibility of extracting spectroscopic information from the fluctuation spectra of radiation transmitted by a medium[1-2] or emitted spontaneously by a medium.[3-4] In both cases, we were concerned with the characteristic transformation of the fluctuation spectrum of an external source by a medium. Thus, in the first case, we considered the transformation of the statistics of radiation absorbed or amplified by a given medium and, in the second case, we discussed the transformation of the fluctuation spectrum of an exciting process into the fluctuation spectrum of spontaneous emission. The transformation of the spectrum of primary noise can be looked upon as the result of an active or passive filtration of this noise by the medium. A necessary condition for the observation of such processes is the avilability of sources of broad-band noise of the necessary intensity.

It is, however, possible to modify investigations of this type whereby a medium under investigation acts not as a filter of externally induced noise but a modulator with a characteristic spectrum. The experimental conditions are then radically altered: whereas in the schemes considered previously, the requirement was, for example, for maximum possible fluctuations, we now require radiation with minimum noise, so that the appearance of fluctuations associated with the interaction with the medium becomes noticeable.

Suppose that the problem is to investigate the dynamics of fluctuations in the number of particles at a certain level $k$. Population fluctuations can be recorded as fluctuations in absorption associated with $k \rightarrow n$ transitions, provided the following obvious conditions are satisfied: (1) the incident radiation must be sufficiently strong in intensity to ensure that the required fluctuations can be recorded against the background of natural (shot) fluctuations of light, and (2) it is desirable for the absorption process to be linear since, otherwise, the fluctuations under investigation will be modified (smoothed) by the action of the incident light. The linearity conditions means that, firstly, there is no optical pumping and, secondly, the rate of induced $k \rightarrow n$ transitions is much smaller than the rate of the reverse transitions.

These requirements can readily be satisfied in studies of slow fluctuations, for example, fluctuations in the number of atoms in a given volume of a vapor-containing vessel. Optical recording of the dynamics of such fluctuations is interesting in connection with problems in diffusion and self-diffusion of gases. On the other hand, fluctuations in absorption could act as sources of excess noise for discriminators used with optical and radio-optical resonances.

2. The situation of fluctuations in the absorption of a gas was discussed in detail in[5]. Here, we confine our attention to a simple estimate of fluctuations in the light flux passing through a cross section $S$ of a gas of length $l$ with mean particle density $\bar{n}_0$ (here and henceforth, a bar will indicate a mean value). Each particle has a probability $Sl/V$ of being in the volume $Sl$ cut by the light beam out of the total volume $V$. The particle distribution is binomial so that, when the number of particles is large, it can be replaced with good approximation by a Gaussian distribution with variance

$$\overline{(N-\bar{N})^2}=\bar{N}(1-Sl/V), \quad \bar{N}=Sl\bar{n}_0.$$

For simplicity, we shall assume henceforth that $Sl/V \ll 1$ and that the variance is equal to $\bar{N}$. Diffusion will ensure a continuous variation in the realization of the distribution with a certain characteristic correlation time $\tau_c$ related to the mean time taken by a particle to leave the volume $Sl$.

Suppose that the intensity of the incident light is $I_0$ photons per second. The intensity of the transmitted light is

$$I=I_0e^{-\sigma N/S} \tag{1}$$

and fluctuates because of the fluctuations in the number $N$ of particles entering the light beam ($\sigma$ is the differential absorption cross section). Using (1) and the Gaussian distribution for the number $N$ of particles, we can readily verify by direct calculations that the mean square fluctuation in the intensity of the transmitted light is

$$\overline{(I-\bar{I})^2}=(\bar{I})^2k^2(\bar{N})^{-1}, \quad \bar{I}=I_0e^{-k}, \tag{2}$$

where $k=\sigma\bar{N}/S$ is the mean gas absorption coefficient. It is assumed in the calculation that $\sigma k/S \ll 1$, which is always true. Assuming that the fluctuation spectrum is approximately rectangular, extending from zero up to frequencies of the order of $\tau_c^{-1}$, we can estimate the spectral density of the fluctuations from the formula

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$$I_\omega^2 = I^2 k^2 \tau_c (\bar{N})^{-1}. \quad (3)$$

To transform from the spectral density to the photocurrent $i_\omega^2$ through the receiver, we must take into account the quantum yield $q$ of the latter:

$$i_\omega^2 = q^2 e^2 I_\omega^2$$

where $e$ is the electron charge. This must be compared with the spectral density of shot fluctuations in the photoelectric current, which is given by the Schottky formula

$$i_{sh}^2 = qe^2 \bar{I}/\pi.$$

The ratio $\eta = i_\omega^2 / i_{sh}^2$ determines the detectability of fluctuations in absorption against a background of shot noise. For frequencies $\omega < \tau_c^{-1}$, this ratio is given by

$$\eta = \pi q \tau_c k^2 \bar{I} (\bar{N})^{-1}.$$

Using the relationship between the absorption coefficient $k$, the number of atoms, and the intensity of transmitted light, we can readily show that the ratio is a maximum for $k = 1$. When this condition is satisfied,

$$\eta = \pi q \tau_c \bar{I} (\bar{N})^{-1} \approx 0.58 \pi q \Delta I \tau_c (\bar{N})^{-1} = 0.58 \pi q \tau_c / T, \quad (4)$$

where $\Delta I = I_0 - \bar{I}$ is the mean number of photons absorbed per second by $\bar{N}$ atoms. The ratio $\bar{N}/\Delta I = T$ is equal to the mean time between successive photon absorptions by the atom.

It is clear from (4) that absorption fluctuations become more appreciable as the frequency with which each particle is excited is increased, i.e., as the light intensity increases and the participation of particles in absorption is reduced (subject to $k = 1$). Absorption fluctuations will therefore increase with increasing absorption cross section $\sigma$ of each particle and decreasing cross section $S$ of a beam of constant intensity, and this makes them particularly important in the case of highly collimated laser beams. It also follows from (4) that the spectral density of fluctuations increases with increasing correlation time $\tau_c$. The fluctuations are then concentrated in the region of increasingly lower frequencies. In practice, the time $\tau_c$ can be varied in a broad range by varying the light-beam cross section and using a buffer gas which slows down the diffusion of absorbing atoms.

In addition to the above component of absorption fluctuations, there is a further component connected with the random redistribution of atoms over the beam cross section with their total number held constant. This component appears as a result of the nonlinear relationship between the intensity of transmitted light and the density of absorbing atoms, which is important for $k \gtrsim 1$. It is shown in[5] that this component has a much lower spectral density for a broader spectrum.

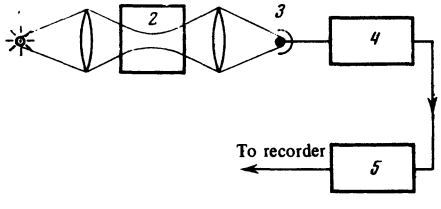


FIG. 1. Experimental arrangement: 1—spectral lamp; 2—absorbing cell; 3—photodiode; 4—selective tunable amplifier; 5—linear detector.

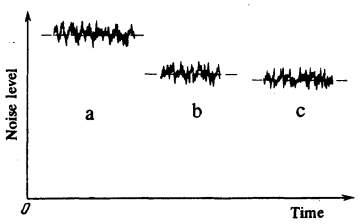


FIG. 2. Typical noise records.

3. The experiment illustrated in Fig. 1 was carried out to detect the above absorption fluctuations. Light from a gas-discharge potassium lamp was collimated into a narrow beam and intercepted by a cell containing saturated potassium vapor at about 60 °C. The cell also contained a buffer gas (argon) at a pressure of 10 Torr. Light transmitted by the cell was passed through an interference filter defining the resonance doublet at 7665 and 7699 Å, and was eventually recorded by the FD-7K photodiode. The photocurrent through the detector was about 50 $\mu$A. The signal taken from the load of the photodiode was amplified by a selective tuned amplifier and was linearly detected with a time constant of 10 sec. It was finally recorded by a strip-chart recorder.

To take into account the presence of possible excess noise in the lamp (i.e., nonshot noise), the cell was periodically replaced by a control cell which was identical with the cell containing the potassium vapor but without the argon buffer gas. The two cells had different correlation times $\tau_c$: in the control cell, this was the time of free path of a potassium atom across the beam caustic, which did not exceed $10^{-4}$ sec, whereas, in the working cell, this time was determined by the diffusion of potassium atoms in argon and was of the order of 0.01 sec. In all other respects, the light transmission conditions were identical in the two cells. By measuring the light broadening of the magnetic resonance line of potassium, we found that the pump time $T$ was of the order of $10^{-3}$ sec. It was therefore expected that the control cell introduced no appreciable fluctuations into the transmitted beam and, therefore, the noise level of this beam was adopted as the reference level.

We also measured the noise level produced by the equivalent illumination of the photodiode with a hot-filament lamp. This noise level was found to be identical with the calculated shot noise and was close to the emission noise of the potassium lamp transmitted through the control cell. The entire installation was carefully vibration-proofed in order to suppress microphonic noise.

Figure 2 shows a typical record of the noise in the intensity of light transmitted by the working cell (a) and

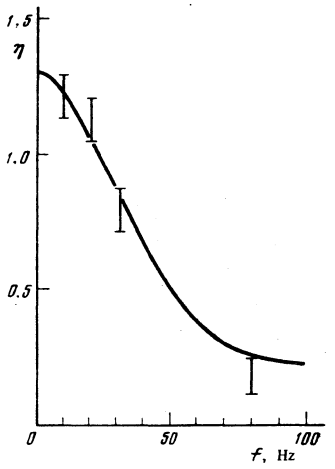


FIG. 3. Frequency dependence of absorption noise level.

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 $\eta$ 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 $\eta$.

4. The experimental data are compared in Fig. 3 with the calculated curve given by the Lorentz formula $I_\omega^2 = I_\omega^2(0)/(\omega^2\tau_c^2+1)$ 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 $\tau_c$. 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 $\tau$ 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

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