Radiative-collisional excitation of sound in methanol vapor

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A new mechanism for the excitation of sound by light in a gas, based on a light-induced drift and a light-induced heat flux, has been observed experimentally for the first time.

In the existing methods for the excitation of sound by electromagnetic radiation, the energy of the sound vibrations is drawn from the energy of a light wave.[1] A fundamentally different mechanism for sound excitation, a "radiative-collisional" mechanism, was proposed in Refs. 2. In this new mechanism, the energy of the sound vibrations is drawn directly from the thermal energy of a gas-filled cell. This mechanism is based on a light-induced drift[3] (LID) and some related effects.

In this letter we are reporting the first experimental observation of sound vibrations excited in a gas by an LID and a light-induced heat flux.

Let us consider the resonant interaction of light with a two-component gas mixture. One component (a) absorbs the light while the other (b) is a buffer, which does not interact with the light. The acoustic vibrations in the gas are described by the following wave equation[2] for the nonequilibrium increment δP in the total pressure (P) of the gas mixture:

$$\left(\frac{\partial^2}{\partial t^2} - c^2\Delta\right)\delta P = -\frac{\partial}{\partial t}\nabla c^2 \frac{n_a n_b}{n}(m_b - m_a)(\vec{u}_a - \vec{u}_b) + \frac{2}{3}\vec{h}] + \zeta, \tag{1}$$

$$\zeta = \frac{\partial Q}{\partial t} + c^2 \sum_{rs} \frac{\partial^2}{\partial x_r \partial x_s}\pi_{rs}.$$

Here c is the sound velocity; $\vec{u}_\alpha$ is the average velocity of component $\alpha = (a, b)$ of the gas mixture, which is equal to the sum of the velocity of the LID[3] and the velocity of light-induced diffusion;[4] $\vec{h}$ and π_{rs} are the light-induced heat flux and the light-induced pressure tensor;[5] and $n = n_a + n_b$, when n_α and m_α are the concentration and mass of the particles of species $\alpha = (a, b)$. In a weak field, the quantities $\vec{u}_\alpha$, $\vec{h}$, and π_{rs} are proportional to the light intensity and to the relative difference between the transport collision rates for the excited and unexcited particles. If the light intensity varies in time, the terms on the right side of Eq. (1) are responsible for an excitation of sound vibrations in the gas by the electromagnetic field. The term $\partial Q/\partial t$ in (1), where Q is the part of the radiant power which is dissipated as thermal energy of the gas, is responsible for the optoacoustic effect.[1] The rest of the right-hand side of (1), which arises from the velocity-selective interaction of the particles with the light,[1)] describes a "radiative-collisional" excitation of sound.[2,6,7] The particle flux $n_\alpha \vec{u}_\alpha$ and the heat flux $\vec{h}$ differ from Q and from the pressure tensor π_{rs} in that they are antisymmetric functions of the detuning (Ω) of the light, at the frequency ω, from the resonant frequency ω_0. This circumstance is crucial for distinguishing the contribution (δP_{LID}) from the light-induced particle and heat fluxes to the total amplitude of the sound vibrations, $\delta P = \delta P_{\text{LID}} + \delta P_T$, against the background of the optoacoustic signal δP_T. From Eq. (1) we find the following estimate for the ratio of δP_{LID} and δP_T (Ref. 2):

$$\eta = \frac{\delta P_{\text{LID}}}{\delta P_T} \sim \left(\frac{\nu}{\nu_k}\right)^2. \tag{2}$$

Here ν is the frequency at which the light intensity is modulated. The "critical" frequency ν_k is related to the LID velocity $\vec{u}_{\text{LID}} \sim \vec{u}_a - \vec{u}_b$ and the heat flux $\vec{h}$ in the following way:

$$\nu_k^{-1} = 2\pi\left|\frac{kT}{\hbar\omega}\frac{l_0}{c\nu_T}\left(\frac{n_b}{n}\left(\frac{m_a - m_b}{m_a}\right)v_{\text{LID}} + \frac{h}{nm_a c^3}\right)\right|^{1/2}, \tag{3}$$

where T is the gas temperature, l_0 is the photoabsorption length, and ν_t is the rate at which the excited state is quenched. We see from (2) and (3) that a radiative-collisional excitation of sound is possible in both two-component and single-component ($n_b = 0$) gases.

The basic part of the experimental apparatus is a resonant optoacoustic cell (Fig. 1). This cell has two channels, for the "forward" laser beam (A) and the "backward" one (B). An MKÉ-3 microphone is placed in the lateral part of one of the channels. Holes 1 mm in diameter in the plastic bottom of the microphone capsule allow pressure equalization on the two sides of the membrane during the evacuation of the gas. These channels are 15 mm long and 3 mm in diameter. The ends of the cell, 7 mm wide, are covered with KBr windows. A tunable waveguide CO_2 laser, with line selec-

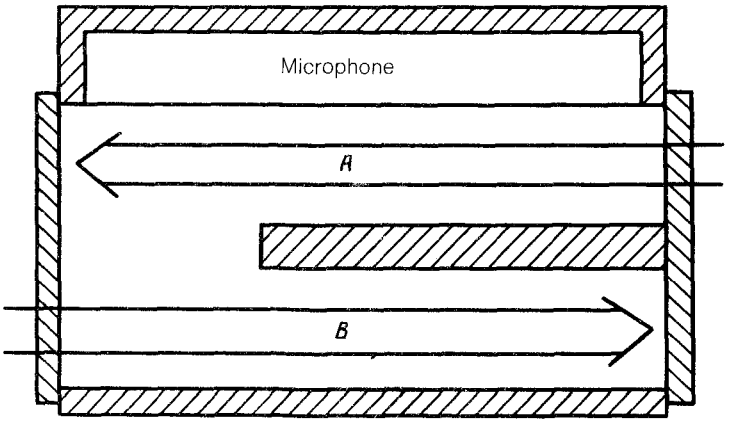

FIG. 1. The optoacoustic cell.

tion by a diffraction grating, is used.[8] The tuning interval is about 300 MHz; the output power is about 1 W. The most suitable line for the purposes of the present study is the line 9P16 of the CO_2 laser; it lies close to the $R(0,0;11)^{CO} \leftarrow (0,0;10)^{O}$ transition in the CH_3OH molecule. The laser light is amplitude-modulated by an ML-7 electrooptic modulator. A Ge beam splitter positioned after this modulator forms the forward (A) and backward (B) beams. A system of mirrors rotates the backward beam into the appropriate channel of the cell, in such a way that the diameters of the forward and backward beams are identical.

The experiments were carried out with methanol vapor with the typical parameter values[8] $u_{\mathrm{LID}} \sim 1$ cm/s, $c = 3.4 \times 10^4$ cm/s, $n_b/n \lesssim 1/2$, and $\nu_T \sim 10^3$ Hz. Plugging these values into (3), we find $\nu_k \sim 50$ kHz. According to (2), this result means that the radiative-collisional mechanism for the excitation of sound outweighs the optoacoustic mechanism in the region $\nu > 50$ kHz. Unfortunately, the sensitivities of the microphones to which we had access fell off sharply at $\nu > 20$ kHz. We accordingly used a different method to suppress the optoacoustic signal.

Sound was excited in the cell at its resonant frequency of 18 kHz. At this frequency, as we see from (2), the radiative-collisional signal is an order of magnitude weaker than the optoacoustic signal ($\eta \sim 10^{-1}$). To suppress the optoacoustic signal, we made use of a particular feature of the 18-kHz resonance. We interpreted this resonance as a Helmholtz resonance,[9] since the experimental value of the frequency agrees approximately with the calculated value $\nu_G = c/2\pi[S/V(l + \pi R/2)]^{1/2} \approx 15$ kHz (more on this below). Here $S = \pi R^2$ and l are the cross-sectional area and length of the neck connecting the channels, and V is the reduced volume of the channels. A Helmholtz resonance corresponds to a periodic movement of gas through the neck, from one channel into the other. When the forward and backward beams (Fig. 1) are directed opposite each other, the absorbing particles and also the heat "flow" from one channel of the cell into the other. The radiative-collisional signal, in contrast with the optoacoustic signal, will be amplified at the frequency of the Helmholtz resonance. This circumstance makes it possible to suppress the optoacoustic signal when the

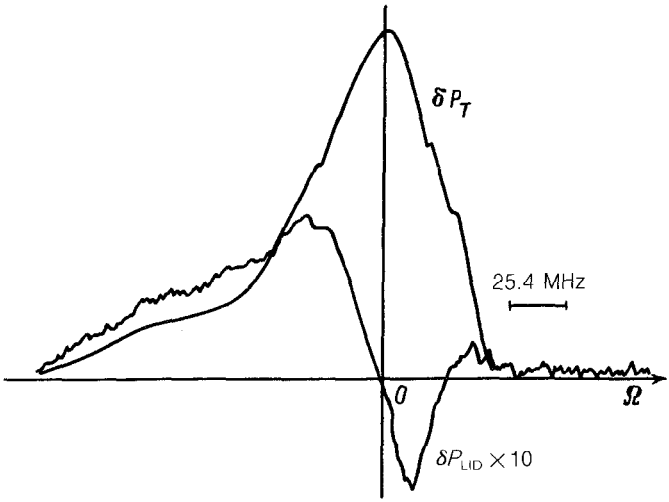

FIG. 2. The amplitudes of the optoacoustic (δP_T) and "radiative-collisional" (δP_{LID}) signals versus the frequency detuning $\Omega = \omega - \omega_0$.

intensities of the forward (A) and backward (B) beams are balanced appropriately at the center of the CH_3OH absorption line. At other resonant frequencies of the cell, it was not possible to achieve this suppression. That result is another strong argument for the conclusion that 18 kHz is the frequency of a Helmholtz resonance.

Figure 2 shows the δP_T absorption lineshape of the methanol vapor, recorded during excitation of the sound by only a single laser beam, along with the acoustic signal δP_{LID} during the excitation of sound by two oppositely directed laser beams (Fig. 1). The acoustic signal δP_{LID} has the spectrum characteristic of LID, i.e., the

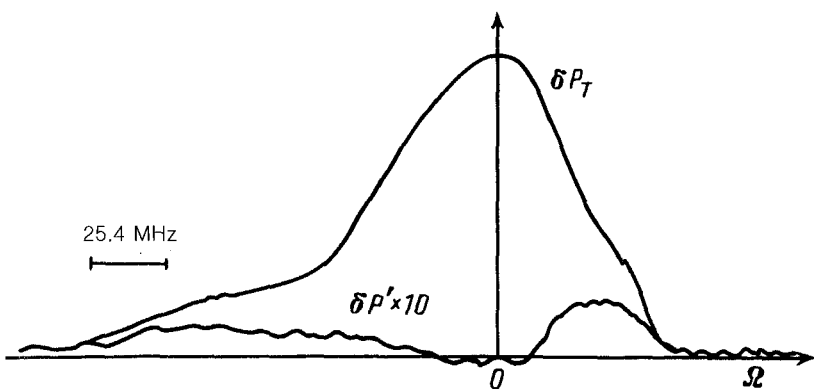

FIG. 3. Amplitude of the acoustic signal, $\delta P'$, versus the frequency detuning Ω for the case in which beams A and B are propagating in the same direction.

signal is proportional to the derivative of the δP_T absorption lineshape with respect to Ω. Figure 2 corresponds to a mixture of CH_3OH and Xe (a 1:1 mixture with a total pressure of 1.5 Torr and with $\eta \sim 0.05$). Similar curves were found for a mixture of CH_3OH with H_2 and for pure CH_3OH. The presence of a signal in the pure CH_3OH agrees with Eqs. (2) and (3), which show that in the case $n_b = 0$ an acoustic signal with an antisymmetric Ω dependence is excited by the light-induced flux of heat or internal energy.

Figure 3 shows the results of a control experiment in which beam *A* propagated in the same direction as beam *B*, in contrast with the case in Fig. 1. As expected, there is no signal which is an antisymmetric function of Ω at the center of the absorption line. The residual signal, which is a symmetric function of Ω near the center of the absorption line, is a consequence of an incomplete suppression of the optoacoustic signal in the case $\Omega \neq 0$ because of the finite photoabsorption length.

[1)] We are ignoring some weaker effects such as light-induced diffusion.[4]

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