Theory of self-oscillation and mode locking in a longitudinal photoacoustic resonator

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The wave equation for pressure that governs generation of the photoacoustic effect possesses a forcing term proportional to the time derivative of the energy delivered to the gas per unit volume and time. A positive pressure fluctuation, with its accompanying density increase, thus increases the optical absorption and provides a positive feedback mechanism for sound generation. A theory for self-oscillation in a one-dimensional resonator is given. Expressions for the photoacoustic pressure are derived for the cases of highly and weakly absorbing gases that indicate mode-locked sound generation. Experiments with CO_2 lasers are reported where evidence of the self-generation effect was sought.

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I. INTRODUCTION

The photoacoustic effect [1–3], which refers to the production of sound by absorption of optical radiation, generally is produced as a result of thermal expansion following the optical deposition of energy. When heat conduction and viscous effects are ignored, the photoacoustic pressure p is governed by the wave equation [4]

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right) p(\mathbf{x},t) = -\frac{\beta}{C_P}\frac{d}{dt}H(\mathbf{x},t), \quad (1)$$

where c is the sound speed, β is the thermal expansion coefficient, C_P is the specific heat capacity, t is the time, and H is the heating function, which describes the energy per unit volume and time delivered by the optical source to the absorbing medium. It is evident from the form of the forcing term in Eq. (1) that sound can be generated by any optical source that provides constant heat deposition but which varies in space [1,5,6], or as in, for example, trace gas detection, for any source that is invariant in space, but which varies in time [7]. It has been recognized by Kolomenskii and Maznev [8] that since an increase in pressure in a propagating acoustic wave is accompanied by a corresponding increase in density, when an optical beam with a wavelength corresponding to an absorption of the gas is present, the density increase leads to an increase in optical absorption and a further pressure increase, so that amplification of the wave is possible—even for the case of a continuous optical beam whose intensity varies neither in time nor in space.

Here, the generation of longitudinal acoustic waves by a continuous laser beam directed into a resonator is considered based on the amplification inherent in the mechanism of sound production by the photoacoustic effect. The theory is formulated for a one-dimensional resonator with plane parallel surfaces, one of which acts as a window for the entrance of a continuous laser beam that is absorbed by an inviscid gas. In the region near the entrance window, where a pressure antinode exists, any pressure increase in a standing wave results in an increased density of absorbers and hence additional energy deposition relative to gas in the cell at the ambient pressure. Correspondingly, when the pressure decreases near the window on the next half cycle of the acoustic standing wave, a smaller amount of energy is deposited relative to gas at ambient pressure. The result is a reinforcement of the pressure amplitude of the standing wave, or, equivalently, an amplification of the standing wave.

Consider a laser beam with a uniform intensity I_0 directed into a cell containing a gas with a density ρ with an optical absorption coefficient per unit density $\hat{\alpha}$, as depicted in Fig. 1. The heating function can be written as

$$H(\mathbf{x},t) = \hat{\alpha}(\rho+\delta)I(\mathbf{x},t), \quad (2)$$

where δ is the acoustic density and I is the optical beam intensity. From linear acoustics [9,10], the density and pressure are related by the relation $\delta = p/c^2$, which, when substituted with Eq. (2) into Eq. (1), gives for a one-dimensional resonator

$$\frac{\partial^2 p}{\partial z^2} + \frac{\bar{\alpha}\beta I_0}{\rho c^2 C_P}e^{-\bar{\alpha}z}\frac{\partial p}{\partial t} - \frac{1}{c^2}\frac{\partial^2 p}{\partial t^2} = 0, \quad (3)$$

where the beam intensity is taken to be absorbed exponentially in space, with an absorption coefficient $\bar{\alpha}$ given by $\bar{\alpha} = \hat{\alpha}\rho$; that is, the effect of a change in pressure the exponential function has been taken to be negligible.

The generation of the acoustic signal in the resonator, with the assumptions noted above, is governed by Eq. (3), which is a partial differential equation in space and time. In Sec. II, the properties of sound generation as governed by Eq. (3) are discussed for a strongly absorbing gas, which shows a mode coupling effect. Section III gives a frequency domain, series solution to Eq. (3) where the amplification effect is described for the general case of an absorption coefficient of arbitrary magnitude. A solution is given for a weakly absorbing using the series solution. Section IV discusses experiments carried out with CO_2 lasers irradiating cells filled with SF_6, and Sec. V, the Discussion section, gives an overview of the self-oscillation effect.

II. SOLUTION FOR A STRONGLY ABSORBING MEDIUM

When the absorption of the gas become high, it is possible to approximate the exponential factor in Eq. (3) through use of the relation

$$e^{-\lambda f(z)} \underset{\lim\ \lambda\to\infty}{=} \delta(z-z_0)\int e^{-\lambda f(z')}dz', \quad (4)$$

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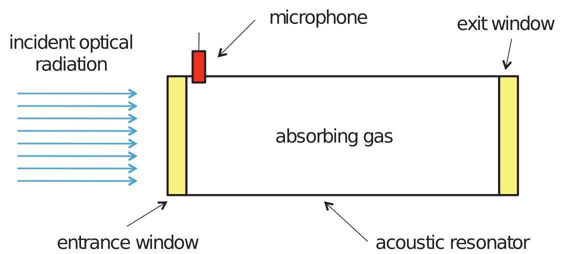

FIG. 1. (Color online) Diagram of a photoacoustic cell for producing self-oscillation in a gas. The radiation enters from the left at a coordinate $z = 0$. In some experiments the microphone was mounted at the exit window; in others it was as shown.

where $\delta(z)$ is the Dirac δ function. With this approximation, and taking the pressure to vary as $\tilde{p}(z)\exp(-i\omega t)$, Eq. (3) becomes

$$\frac{\partial^2 \tilde{p}}{\partial z^2} + k^2\tilde{p} = \frac{i\omega\beta I_0}{\rho c^2 C_P}\tilde{p}(0)\delta(0). \tag{5}$$

A Green's function for the resonator that has boundary conditions such that the acceleration in the acoustic wave $\nabla p/\rho$ is zero at $z = 0$ and L can be found [11] to be

$$G_A(z,z') = \frac{1}{\sin kL}\begin{cases}\cos kz \cos k(z' - L) & z < z' \\ \cos kz' \cos k(z - L) & z > z'\end{cases}. \tag{6}$$

The Green's function solution to a Helmholtz equation of the form of Eq. (5) with a source function $S(z)$ is given by

$$\tilde{p} = \int G(z,z')S(z')dz'. \tag{7}$$

The integration of the Green's function over the source term in Eq. (5) is trivial as the integral $\int_0^L \cos kz'\delta(z')dz'$ is unity; thus, the time domain acoustic pressure becomes

$$p(t) = -\frac{\beta I_0 \tilde{p}(0)}{2\pi c\rho C_P}\int_{-\infty}^{\infty}\frac{e^{-i\omega[t-(z-L)/c]} + e^{-i\omega[t+(z-L)/c]}}{e^{ikL} - e^{-ikL}}d\omega, \tag{8}$$

which can be written

$$p(t) = \frac{\tilde{p}(0)}{2\pi}\hat{\Gamma}\sum_{n=0}^{\infty}\int_{-\infty}^{\infty}[e^{-i\omega[t-(z-L)/c]} + e^{-i\omega[t+(z-L)/c]}] \times e^{2i(\omega L/c)n}d\omega, \tag{9}$$

where

$$\hat{\Gamma} = \frac{\beta I_0}{c\rho C_P}.$$

Each term in the series given by Eq. (9) can be evaluated as a δ function so that Eq. (9) becomes

$$p(t) = \tilde{p}(0)\hat{\Gamma}\sum_{n=0}^{\infty}\left[\delta\left(t - \frac{z + (2nL)}{c}\right) + \delta\left(t + \frac{z - (2n+2)L}{c}\right)\right]. \tag{10}$$

Since on every reflection of the acoustic pulse at the entrance window the pressure is augmented by a factor of Γ, Eq. (10) must be modified to give

$$p(t) = \tilde{p}(0)\sum_{n=0}^{\infty}\hat{\Gamma}^{n+1}\left[\delta\left(t - \frac{z + (2nL)}{c}\right) + \delta\left(t + \frac{z - (2n+2)L}{c}\right)\right], \tag{11}$$

which shows a series of pressure transients that increase with each pass through the resonator. The δ functions travel within the boundaries of the cell and are considered to exist within the photoacoustic cell only. Initially, the right-going δ function, corresponding to $n = 0$, leaves the left window at $z = 0$ where the laser beam enters, travels inside the cell to the point $z = L$, and exits, at which time the left-going δ function launched at $z = 2L$ arrives at $z = L$, and propagates inside the cell. When this latter pulse reaches the point $z = 0$ the right-going pulse from the $n = 1$ term enters the cell. When all of the terms are considered, a traveling δ function pulse that propagates continuously back and forth inside the cell is described.

III. SERIES EXPANSION SOLUTION

When Eq. (3) is written so that all quantities vary as $\exp(-i\omega t)$, the wave equation for pressure becomes

$$\frac{\partial^2\tilde{p}}{\partial z^2} + k^2\tilde{p} = \Gamma e^{-\alpha z}\tilde{p}, \tag{12}$$

where

$$\Gamma = \frac{i\omega\alpha\beta I_0}{\rho c^2 C_P}.$$

A series expansion solution for the pressure can be obtained by considering the right hand side of Eq. (12) to be of order ε, and expanding the pressure in a series of functions f_j as

$$p = \varepsilon f_1 + \varepsilon^2 f_2 + \varepsilon^3 f_3 + \cdots. \tag{13}$$

Substitution of Eq. (13) into Eq. (12) and equating terms with identical powers of ε gives each f_j as solutions to

$$\begin{aligned} f_1'' + k^2 f_1 &= 0, \\ f_2'' + k^2 f_2 &= \Gamma\, f_1 e^{-\bar{\alpha}z}, \\ f_3'' + k^2 f_3 &= \Gamma\, f_2 e^{-\bar{\alpha}z}, \\ f_j'' + k^2 f_j &= \Gamma\, f_{j-1}e^{-\bar{\alpha}z} \quad \text{for arbitrary } j. \end{aligned} \tag{14}$$

The solution for the first of these that satisfies the boundary conditions at the ends of the cell would be a superposition of the eigenmodes of the cavity, $p = \sum \tilde{p}_n \cos k_n z$, where $k_n = n\pi/L$.

Consider the solution of Eq. (14) for a single eigenmode $\tilde{p}_m \cos k_m z$. The solution for f_2 from the second of Eq. (14) can be found using a Green's function of the form

$$G_B(z,z') = \frac{2}{L}\sum_n \frac{\cos k_n z \cos k_n z'}{k^2 - k_n^2} \tag{15}$$

to give a first approximation to the photoacoustic pressure as

$$p = p_m\left[\cos k_m z + \frac{2}{L}\Gamma\sum_{n,m}\frac{\cos k_n z}{k^2 - k_n^2}I_{nm}\right], \tag{16}$$

where

$$I_{nm} = \int_0^L \cos k_n z \cos k_m z \, e^{-\alpha z} dz. \tag{17}$$

Although the integral in Eq. (16) can be evaluated analytically, solutions to the subsequent equations for f_3 or higher result in complicated, lengthy expressions.

Weakly absorbing gas

For the case where the absorption coefficient is small, the exponential function in the expression for I_{nm} can be approximated as unity so that $I_{nm} = L\delta_{nm}/2$, where δ_{nm} is the Kroneker δ function. The solution for f_2 using Eq. (7) with G_B gives

$$f_2 = p_m \Gamma \frac{\cos k_m z}{k^2 - k_m^2}. \tag{18}$$

Using f_2 as a source, according to the third of Eq. (14), gives

$$f_3 = p_m \Gamma^2 \frac{\cos k_m z}{\left(k^2 - k_m^2\right)^2}. \tag{19}$$

It is not difficult to show that for the general term f_q the solution is

$$f_q = p_m \Gamma^{q-1} \frac{\cos k_m z}{\left(k^2 - k_m^2\right)^{q-1}}. \tag{20}$$

The series given by Eq. (13) is of the form of of a power series $1 + x + x^2 + x^3 + \cdots = (1 - x)^{-1}$, where $x = \Gamma/(k^2 - k_m^2)$; thus, the series can be summed to give the acoustic pressure as

$$\tilde{p} = \tilde{p}_m \cos k_m z \left(\frac{k^2 - k_m^2}{k^2 - k_m^2 - \Gamma} \right). \tag{21}$$

If Eq. (21) is Fourier transformed into the time domain, the photoacoustic pressure becomes

$$p(z,t) = \frac{\tilde{p}_m \cos k_m z}{2\pi} \int_{-\infty}^{\infty} \frac{\omega^2 - (ck_m)^2}{(\omega - \omega_-)(\omega - \omega_+)} e^{-i\omega t} d\omega. \tag{22}$$

The poles in Eq. (22) are found for small Γ_ω to be

$$\omega^{\pm} = \frac{i\Gamma_\omega}{2} \pm ck_m,$$

where

$$\Gamma_\omega = \frac{\bar{\alpha}\beta I_0}{\rho C_P}.$$

It is convenient to write Eq. (22) using the operator $-[d^2/dt^2 + (ck_m)^2]$, which reduces the numerator in the fraction in the integral to unity. As both poles lie in the lower half complex ω plane, the integration is straightforward giving

$$p(z,t) = \frac{\tilde{p}_m \cos k_m z}{ck_m} \left[\frac{d^2}{dt^2} + (ck_m)^2 \right] \times \sin\left[ck_m \left(1 - \frac{\bar{\Gamma}^2}{(2ck_m)^2} \right)^{1/2} t \right] e^{(\bar{\Gamma}/2)t}. \tag{23}$$

On evaluating Eq. (23) to first order in Γ_ω, the photoacoustic pressure is found to be

$$p(z,t) = \tilde{p}_m e^{(\Gamma_\omega/2)t} \cos k_m z \cos ck_m t, \tag{24}$$

which describes a standing wave growing in amplitude at a rate $\Gamma_\omega/2$ in time.

IV. EXPERIMENTS

Experiments seeking evidence for the generation of sound by absorption of continuous laser radiation were conducted over a period of time using four different CO_2 lasers. The 10.6-μm beams from the lasers were directed into several different resonators whose lengths were 11, 17, or 37 cm long, with inside diameters of 12.5 mm or smaller. The cells were filled in different experiments with various mixtures of SF_6 in N_2 with mole fractions of 0.05, 0.08, 0.09, and 1. Note that the largest signals were expected at approximately 90% SF_6 owing to a heat conduction effect where optically thick gases transmit heat to the entrance window of the photoacoustic cells resulting in acoustic signal diminution, as has been previously reported [12]. The acoustic signals were recorded with a condenser microphone (B & K Inc., Model 4130) whose output was viewed on a digitizing oscilloscope.

In all of the experiments carried out, there was little difficulty in seeing an acoustic signal at either the fundamental or one of the overtones of the longitudinal resonance frequency of the cell as long as the laser power exceeded roughly 0.2 W. The difficulty in ascertaining that self-oscillation was in fact taking place was that oscillation could be excited by transients that arose from plasma oscillations in the laser, or, more typically, from small voltage spikes that were generated by the power supply, which gave transient power fluctuations on the laser outputs. Even with power supplies based on full wave rectification of 60-Hz high voltage, transients were found at the line frequency which arise from nonideal behavior of the high voltage diodes in the voltage rectifier circuit. When transients in the laser power are generated in this way, it is easy to distinguish between self-oscillation and sound generation by transients by triggering the oscilloscope on the line voltage, and signal averaging the microphone waveform. A clear sign that the sound does not arise from self-oscillation is that the amplitude of averaged signal even though it appears at a longitudinal resonance of the cell averages to a finite amplitude synchronous with the line frequency. Self-oscillation will not be synchronous with the line frequency and the waveform should decay to zero after multiple averages.

In order to reduce the amplitudes of the transients in the power supply for the flowing gas laser (Advanced Kinetics Inc., Model MIRL 50), a single stage *RC* circuit using high voltage capacitors was used. As this proved to be only marginally successful in reduction of the transients, a feedback circuit employing a liquid nitrogen cooled HgCdTe infrared detector was used to stabilize the power supply through the laser's external voltage control unit. As this proved inadequate for suppressing the transients, a sealed CO_2 laser (Parallax Tech Inc.) powered with a high voltage supply (Unipower Inc. Model BRC-30-25P-PX50) that employed a switching supply to provide the high voltage was used. Although no transients at the switching frequency of 40 kHz were detected, again, 60-Hz transients were found on the laser output. To further stabilize the power supply, an Agilent Inc. Model 6035A supply with a ripple specified as less than 0.05% was used to replace the first stage of dc power generation that fed the switching circuit in the laser power supply. This modification of the laser power supply resulted in transients on the laser output on the order of 3% of the continuous output power. Later, for even better stability, the Agilent supply was replaced by 15 lead acid storage batteries wired in series that gave the 180 V required by the switching circuit. Additionally, a feedback circuit comprising an acousto-optic modulator and a HgCdTe infrared detector was employed to reduce the amplitude of the transients and stabilize the laser power further.

With the last modification of the laser power supply, experiments were carried out at powers up to a maximum of 6 W, where no detectable transients on the laser output were found, as determined by the HgCdTe detector. However, no self-oscillation at this power level was found with any of the resonators filled with SF_6-N_2 mixtures that ranged over the percentages noted above.

V. DISCUSSION

At this point, the experiments reported here can only be used to put a bound on the power necessary to generate self-oscillation. It is possible that a highly stable laser with higher output power, or possibly the employment of a spherical resonator, which can be expected to have a higher quality factor than a cylindrical resonator, would uncover the self-oscillation. The remarkable capability of the photoacoustic effect for trace detection at the sub ppm level serves as a strong indicator of a corresponding high sensitivity of the photoacoustic effect to small fluctuations in optical power when a strongly absorbing gas is present in the resonator.

The calculations given above do not give the effects of losses in the cavity which act to damp out self-oscillation. Such losses can be incorporated into the above results by considering the cavity quality factor Q_n the value of which is given a subscript indicating a dependence on the longitudinal oscillation mode number n. For the calculation leading to a single mode, standing wave pressure, the exponential term in Eq. (24) must be replaced by $\exp[\Gamma_\omega - (\omega_n/Q_n)](t/2)$, where ω_n is the angular frequency of the the mode n. For Eq. (11), the incorporation of losses is complicated in that each δ function contains a wide spectrum of frequencies corresponding to a sum over all of the longitudinal modes of oscillation, the damping for each mode on a single cycle being $\exp(-2\pi n/Q_n)$. If the simple case of a frictional force proportional to the wave speed at the wall is taken [9], then the quality factor becomes proportional to n so that $\hat{\Gamma}$ becomes $\hat{\Gamma} = [(\beta I_0/c\rho C_P) - \exp(-2\pi/Q)]$. It is to be noted that the effects of dispersion, which have not been included in the calculations given here, would degrade the sharply spiked pressure profile indicated by Eq. (11); strong deviation from the δ function pressure profile would be expected in an experimental realization of the mode locking effect. The question of what factors limit the amplitude of the photoacoustic wave once oscillation has been attained is not addressed in the formulation given here, but can be found by considering the effects of nonlinear acoustics as discussed in Ref. [8].

The results given here are carried out assuming an initial pressure $\tilde{p}$ that is amplified on interaction with the optical beam, which provides a straightforward starting point for calculations. Although self-oscillation is perhaps excited most easily by an external perturbation, at all times there is thermal excitation of the various modes of oscillation in the cavity depending on their energy relative to $k_B T$, where k_B is Boltzmann's constant and T is the ambient temperature. It is thus possible that self-oscillation can take place spontaneously on achieving a sufficiently high gain in the resonator.

Any single mode can sustain oscillation independently of the other modes since the energy deposition increment (or decrement) is always in phase with the pressure. The salient result given by Eq. (11), however, is that the photoacoustic effect from a continuous optical source causes a locking of the various longitudinal modes of oscillation, the mechanism of synchronizing the modes being a pressure increase at the entrance window in a given mode that increases the pressure in the other modes. As any pressure increase at the entrance window results in increased absorption of energy from the optical beam, the modes are naturally excited to reinforce each other so that phase matching is inherent in the excitation process. This mode locking, under ideal circumstances, would result in the train of sharp pulses described by Eq. (11), which would appear to be directly analogous to the pulse train seen in a mode locked picosecond laser. Despite the apparent similarity between photoacoustic and mode locked laser generation of pulses, the photoacoustic process does not require imposition of a periodic loss in the oscillator. Photoacoustic generation of sound according to the theory given here arises from energy deposition in the wave dependent on the absorbed optical power and acoustic wave amplitude, and appears to be unique in its mechanism of mode locking.

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