An Analytic Description of Light Emission in Sonoluminescence

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Light emission in sonoluminescence is shown to be a lasing process with a wide gain bandwidth. Population inversion of the gas molecules inside the bubble is achieved by hydrodynamical pumping. Analytic expressions are derived for the sonoluminescence pulse: shape, pulsewidth, and their codependence on the spectrum and intensity in physically relevant regimes. A detailed comparison with experiments (R. Hiller et al., Phys. Rev. Lett. 80, 1090 (1998); M.J. Moran et al., Phys. Rev. Lett. 80, 4987 (1998); R. Pech et al., Phys. Rev. Lett. 81, 717 (1998)) suggests excellent agreement.

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The phenomenon of sonoluminescence (SL)—emission of a short pulse of light in an acoustically levitated bubble during its final stage of compression—has attracted a lot of interest [1]. Unfortunately, there does not exist a complete theory that formally explains all aspects of the emitted light [2,3]. Light emission in SL can be understood without invoking bubble dynamics in contrast to the recent claims [4] that a modified bubble dynamics theory could numerically account for the nature of the pulse [2,3]. We argue that population inversion in the medium of gas molecules, which must contain metastable states, generates the SL pulse; towards that end, we explicitly calculate measurable pulse characteristics and compare with the experimental observations [4,5]. However, bubble dynamics is important in evaluating the temperature inside the bubble with precision from ambient parameters such as pressure, bath temperature and equilibrium radius.

We argue that light emission in SL results [6] mainly due to the following two steps: (i) excitation of both gas atoms and molecules containing metastable states at a rate high enough to sustain population inversion against dephasing and loss, (ii) spontaneous and stimulated emission of the excited atomic/molecular levels, collective due to a high density. We consider a realistic model of an argon gas bubble and evaluate various parameters of the emitted light pulse in view of the proposed mechanism of sonoluminescence as a broadband laser [2,3]—though an inefficient one due to the presence of strong dephasing and loss. In this exercise of SL from an argon bubble [3], we derive explicit analytic expressions for (i) the temporal shape of the pulse in various relevant regimes, (ii) the pulsewidth consisting of the rise time—denoting the growth of the pulse, and the fall time—denoting the decay, both compared with recent streak camera experiments in ref. [4,5]; (iii) their individual dependence on wavelength, compared with experiments in ref. [6,7]; (iv) the intensity of the emitted light and its dependence on the pulsewidth, compared with the experiments in ref. [3], and (v) the spectrum at different ranges of wavelength, compared with experiments in ref. [6,7]. Our results are in excellent agreement with these experiments [2,3].

Chemical Requirement: It is known that the gas temperature inside an argon gas bubble can be as high as 10,000 K to 40,000 K (0.86-3.5 eV) due to hydrodynamic collapse [10]. Heating to such a high temperature is sufficient to cause electronic excitation of argon gas atoms. This process is necessary in creating population inversion in atoms with metastable states essential to standard lasing process. However, for hotter bubbles of argon and other rare gases [11] with higher atomic number (smaller ionization potential) electronically bound excited molecular states are formed, though the interaction between the constituent atoms is mostly repulsive [12]: Ar + Ar* → Ar2*, which then decay to ground state by emitting a photon: Ar2* → Ar+Ar+hn. These excimer states are dissociative in the electronic ground state. Spontaneous and stimulated decay of such molecular excited states describe the underlying physical mechanism of excimer laser systems such as Ar2* or Xe2* lasers. Excimers with strongly repulsive ground states emit broad continuum radiation whereas the weakly bound excimers typically display a radiation spectrum with rotational and vibrational structures, which gets homogeneously broadened at high pressure. The binding energy of the excimers depends on potential-energy curves which are extremely complex, including families of closely nested curves of excited states as well as level crossings linking specific levels. After excitation, the system evolves from the top to the bottom of these curves by relaxing through a nested sequence of electronic states. The chemical kinetic ordering originating from the particle interactions of rare gases is such that no direct thermal path is available, and the only accessible decay path is radiation. We argue that this kind of radiation is what gives the light in sonoluminescence.

Let us now consider the chemical kinetics of argon inside the bubble specifically for this exercise. During the compression of the bubble at high pressures (P ≥ 10^3 atm) and temperatures (kBT_c ~ 3.5 eV), argon is excited (e− + Ar → Ar∗ + e−) or ionized (e− + Ar → Ar^+ + 2e−) with a population of e−E_{ion}/k_BT_c or e−E_{ex}/k_BT_c respectively. Since the ionization energy of argon is ~ 12 eV, this corresponds to a small degree (~ 3%) of ionization at a temperature in the range of 0.86-3.5 eV. The atomic excitation to Ar* is much more efficient. As we will see, a large amount of initial ionization is not important for the formation of excimer states. Three-
body recombination reactions rapidly dimerize the excited atoms (Ar* + 2Ar → Ar2* + Ar) or ionized states (Ar+ + 2Ar → Ar2+ + Ar) [3]. Argon excimers formed mostly in many excited states quench to lower states at high pressures [2] and the excitation resides mostly in the lowest excited levels of atomic and molecular levels. The radiative lifetime \( T_1 \) of the most relevant excited state Ar2* (for the 1u molecular state which dominates over the 0* state) is known [3] to be \( 3 \times 10^{-8} \) s.

Presence of a small amount (\( \ll 10\% \)) of a heavier rare gas in a large amount of a lighter rare gas [8] results in Penning (Ar* + Xe → Ar + Xe* + e-) or associative (Ar* + Xe → ArXe* + e-) ionization; these reactions are very fast and occur at every collision. If Penning or associative ionization is not energetically favored, then the energy of the lighter species transfers to the heavier species by reactions Ar* + Xe → Ar + Xe*, and Ar2* + Xe → 2Ar + Xe* [14].

This roughly describes the processes through which argon gas excimers are formed inside a sonoluminescing bubble. In the same spirit, one anticipates other rare gas, rare-gas-halogen excimers and quasi-bound metal vapor excimers [8] to show lasing in the SL configuration. The above chemical kinetics analysis applies to all other kinds of excimers, though the details of the collision mechanisms and reaction rates are different. In the following we discuss the second step of the process which gives rise to the light emission in SL from an argon bubble: decay of the excimer states Ar2*.

Hamiltonian: Let us define a transition operator \( R_{1}^{ab} \) from any state \(|a\rangle \) to \(|b\rangle \) of the i-th molecule: \( R_{i}^{ab} |a\rangle_{i} = |b\rangle_{i} \). The hamiltonian of the molecular system comprising of multiple levels can then be described by \( H_{ad} = \sum_{i} \sum_{e} \sum_{g} \sum_{r} h(\omega_{e} + \omega_{r}) R_{i}^{ge} \delta R_{i}^{re} \), where the index \( i \) runs over all the molecules, and the index \( e \) and \( g \) define the energy of a given excited state \(|e\rangle \) and ground state \(|g\rangle \), with respect to an average energy \( \bar{\omega}_{0} \). \( \bar{\omega}_{0} \) is chosen, without any loss of generality, to be the difference in energy between the lowest excited state and the lowest ground state. The hamiltonian of the field is given by the sum over all modes \( k \) and polarization \( \alpha \) such that \( H_{rad} = \sum_{k} \sum_{\alpha} h(\bar{\omega}_{k}) \delta R_{k}^{g} \delta R_{k}^{e} \). The interaction term between the molecular states and the field modes is \( H_{int} = \sum_{k} \sum_{\alpha} \sum_{\alpha'} \delta R_{k}^{ge} \delta \alpha \times \delta \alpha' \cdot \gamma_{\alpha} \cdot \gamma_{\alpha'} \) and the polarization dipole matrix element \( \delta R_{k}^{ge} \) along the polarization direction as \( \omega_{k} \), we define the following: \( P^{\pm}(\omega, r, t) = \sum_{i} d_{\omega} R_{i}^{\pm}(r - r_{i}, \omega, t) \). \( N(r, \omega, t) = \sum_{i} R_{i}^{(3)}(r - r_{i}, \omega, t) \) is the number of excited molecules at a transition frequency \( \omega \).

Maxwell-Bloch Equations: The common electric field created by the excited molecules is related to the polarization by the Maxwell’s equation:

\[
\frac{\partial^{2} E^{\pm}(r, t)}{\partial t^{2}} - c^{2} \nabla \times \nabla E^{\pm}(r, t) = -\frac{1}{\varepsilon_{0}} \frac{\partial^{2} P^{-}(r, t)}{\partial t^{2}}. \tag{1}
\]

From the Heisenberg’s equation of motion for \( N \) and \( P \):

\[
\frac{\partial N}{\partial t} = \frac{i}{\hbar} E^{-} \cdot [P^{-} - P^{+}] + \frac{i}{\hbar} [P^{-} - P^{+}] \cdot E^{+}
\]

\[
\frac{\partial P^{+}}{\partial t} = \frac{i}{\hbar} P^{+} + 2i \frac{\hbar^{2}}{\varepsilon_{0}} \cdot \gamma_{\alpha}(E^{-}N + NE^{+}), \tag{2}
\]

which define the Maxwell-Bloch equations (MBE) in operator form [3]. Electric field and polarization in an arbitrary direction, the direction of the detector \( \hat{z} \) for example, can be expressed in the slow-varying approximation in a complex form as \( E^{+}(r, t) = \mathcal{E}(z, t)e^{i(\omega_{0}t - kr)} \) and \( P^{+}(r, \omega, t) = \mathcal{P}(z, \omega, t)e^{i(\omega_{0}t - kr)} \), where \( t \) is replaced by the retarded time \( t - r/c \), introduced for convenience, and \( \mathcal{E} \) and \( \mathcal{P} \) are the slow-varying envelopes.

To describe the effect of dephasing and loss [13], \( \kappa \) denotes propagation loss of the electric field and is related to the refraction index \( \eta \) by \( \kappa c / \eta = \kappa c / \eta \). \( 1/T_{2} \) is the dephasing rate of polarization, \( 1/T_{1} \) is the spontaneous emission rate, \( \Lambda_{p} \) is a noise source of polarization due to the background field, and finally, \( \Lambda \) describes the population inversion by the excitation due to hydrodynamic pumping. Eqs. (1) and (2) now take the following form:

\[
\frac{\partial E^{\pm}(r, t)}{\partial z} = -\kappa E^{\pm} + \frac{i \omega_{0}}{2 \varepsilon_{0} c} \int_{\omega_{0} - \Delta \omega/2}^{\omega_{0} + \Delta \omega/2} \mathcal{P}^{-}(r, t, \omega) g(\omega) d\omega.
\]

\[
\frac{\partial P^{+}(r, \omega, t)}{\partial t} = \Lambda_{p} - \frac{1}{T_{2}} \cdot \mathcal{P}^{+} + 2i \frac{\hbar^{2}}{\varepsilon_{0}} E^{-} \mathcal{N}(r, \omega, t),
\]

\[
\frac{\partial N(r, \omega, t)}{\partial t} = \Lambda - \mathcal{N}(r, \omega, t) \frac{1}{T_{1}} - \frac{i}{\hbar} \mathcal{P}^{+} E^{+} - E^{-} \mathcal{P}^{-}. \tag{3}
\]

The first equation describes the electric field created by the polarization of the molecular excitations of the relevant manifold of states minus the loss of the field due to propagation. \( \Lambda_{p} \) represents the noise polarization due to population inversion and the blackbody radiation, which act as an equivalent input field to stimulate the decay.

The effect of Doppler broadening relevant at high temperatures is represented by the term \( i k v \mathcal{P}^{+} \). In fact, it is not necessary to account for all these phenomenological factors to describe the essential features of the recent SL experiments. To that end, we shall obtain the equation for the radiated pulse for a simplified case in the time domain, by replacing the operators \( \mathcal{E}, \mathcal{P} \) and \( \mathcal{N} \) in the MBE by their complex functional forms. From the first MBE and its complex conjugate, after multiplying by \( e^{\omega_{0}A} \mathcal{E}^{*} \), an expression for the radiated intensity \( I \) is found:

\[
\partial I / \partial z = -2 \kappa I + (\omega_{0} A/2) 2 \text{Im}[\mathcal{E} \int \mathcal{P}(r, \omega, t) d\omega], \tag{4}
\]
where $A$ is the area of the cross-section perpendicular to the direction $z$. If $\mathcal{P} du$ gives a polarization at a central frequency $\omega_0$ averaged over an effective bandwidth $(\Delta \omega)_{eff}$. Differentiating with respect to $t$ and using Eq. (3) one obtains:

$$\frac{\partial I}{\partial t} = - (1 + 2 \kappa L) \frac{I}{T_2} + \frac{2 \omega_0 A d^2 N L}{\hbar} I + \omega_0 A L \rho \mathcal{E}, \quad (5)$$

where $L$ is a characteristic length scale of the active medium. The first term on the right hand side describes dephasing or inhomogeneous broadening $(1/T_2)$ and propagation loss ($\kappa$). The second term denotes the source, and it can be written as $\frac{3 \Lambda}{4 \pi^2} N L T_2 I$. The third term is the polarization source for the input field that drives the decay:

$$\lambda \mathcal{E} = \frac{1}{\omega_0 A L} \frac{\hbar \omega_0}{T_R T_2^{\text{eff}}} \frac{n_2}{n_2 - n_1} + e^{i \omega_0 / \kappa L T - 1}, \quad (6)$$

The effective dephasing time $T_2^{\text{eff}}$ for a gain medium is $T_2^{\text{eff}} = T_2 (2 \alpha_0 L / \pi)^{1/2}$, where $\alpha_0 L = T_2 / T_R$ is the gain over the length $L$, and $T_R (\omega_0, \Delta \omega_{eff})^{-1} = N (\omega_0, \Delta \omega_{eff}) / T_1$ is the collective decay rate at $\omega_0$. The second term gives polarization due to blackbody radiation interacting within $\Delta \omega_{eff} = 1 / (2 T_2)$, and the first term inside the bracket is the excitation spectrum of the population inversion $g (\omega_0, \Delta \omega_{eff}) = n_2 / (n_2 - n_1)$. In thermal equilibrium $n_2 = n_1 e^{-\hbar \omega_0 / \kappa L T}$, thus the right hand side of Eq. (6) vanishes, implying the absence of population inversion.

Coupled equations for intensity and population are obtained from Eq. (5) and (6):

$$\frac{\partial I}{\partial t} = -(1 + 2 \kappa L) \frac{I}{T_2} + \frac{3 \Lambda^2 e}{4 \pi^2} N L \frac{T_1 I}{T_2} + \frac{\hbar \omega_0}{T_R T_2^{\text{eff}}} [g (\omega_0, \Delta \omega_{eff}) + \frac{1}{e^{i \omega_0 / \kappa L T} - 1}], \quad (7)$$

$$\frac{\partial N}{\partial t} = - \frac{N}{T_1} + \Lambda A L - \frac{2 I}{\hbar \omega_0} (1 + 2 \kappa L), \quad (8)$$

$T_1 \equiv T_2$ if $T_2 \ll T_1$ as in the case of SL, else $T_1 \equiv T_1$. **Pulse Shape in an Ideal Case:** In an idealistic case one neglects dephasing, $T_2 = 0$, $\kappa = 0$, and $\Lambda, \rho = 0$, retaining the effects in the initial conditions. We define a time- and space-dependent tipping angle $\theta (z, t)$ in terms of the population and polarization in the medium so that $N = n_0 \cos \theta (z, t)$, and $\mathcal{P} = i d n_0 \sin \theta (z, t) e^{i \omega_0 t}$. The Bloch equations at a given $\omega$ reduce to $(2d / \hbar) \mathcal{E} = e^{i \omega_0 t} \partial \theta / \partial t$, and the Maxwell’s equation to $\partial^2 \theta / \partial z \partial t = (\Gamma N / L_0) \sin \theta$, where $\Gamma N = T_1 N = T - 1$, and $L_0$ is the length scale over which the polarization contributes to the electric field coherently. The MBE can be written as

$$\theta''(u) + \theta' (u) / u - \sin \theta = 0, \quad (9)$$

in a dimensionless variable $u = 2 (T_1 / T_2) \sqrt{T_2}$, demonstrating clearly the natural emergence of a collective timescale $T_R$. Solutions to the damped pendulum equations of this type, known as sine-gordon equations, include a wide range of explicit functions with the constraint $\theta(0) = \theta_0$, the input noise field, and $\theta'(0) = 0$. If $L \ll L_0$, then the entire active medium is coherent and one obtains pure superradiance. Solution of the equation in that limit with the first term omitted yields an expression for intensity $I \propto \sin^2 (t - T_2 / T_R)$, where $T_2 = T_R \ln (N)$ is the pulse rise time $\theta$. When $z > L_0$, then one expects the occurrence of several maxima (ringing) in intensity. The field emitted by molecules in the central coherent volume ($z < L_0$) are reabsorbed by the molecules outside this regime ($L_0 < z < R_{\text{min}}$) to give stimulated emission. Hints of this behavior may have been experimentally observed.

**Pulse Rise Time:** The pulse rise time is defined as the time at which the first peak is observed from the time when the sample is completely inverted. In the small angle limit ($\sin \theta \approx \theta$), Eq. (10) has the standard Bessel function solution of the linearized sine-gordon equation:$\theta (\vartheta) = \theta (0) J_0 (i \vartheta)$. Using the asymptotic limit, $J_0 (i \vartheta) \sim e^{i \vartheta} / \sqrt{2 \pi \vartheta}$, and requiring $\theta (\vartheta, t = T_D) = 1$ for the maximum reached at time $T_D$, one obtains a transcendental expression: $T_D = (T_R / \pi) [\ln (\theta (0)) - (1 / 4) \ln (T_R / 16 \pi^2 T_D)]^2$. The second term inside the bracket is negligible compared to the first for the experimentally relevant parameters, and $T_R / 16 \pi^2 T_D < 10$. Using Eq. (10) one finds $\theta (0) = (2d / \hbar) T_R \sin \theta (0) \propto (\hbar \omega_0) + (\hbar \omega_0 / \kappa L T - 1)^{-1}$. Thus in the range where the excitation spectrum is thermal, $(e^{i \omega_0 / \kappa L T - 1} \gg \hbar \omega_0)$, $T_D = (T_R / \pi) [\ln (k_B T / \hbar \omega_0)]^{-2}$ constant, for $k_B T \gg \hbar \omega_0$, and $T_D = (T_R / \pi) (\hbar / 2 k_B T)^2$ constant, for $k_B T \ll \hbar \omega_0$. In the nonthermal regime of the excitation spectrum, the rise time is independent of temperature. In experiments, since $T$ is usually much larger than $\hbar \omega_0 / k_B$ ($\sim 2500-11000 K$ in the visible range), dependence of $T_D$ on $\lambda$ and $T$ is expected to be logarithmically slow, or almost absent, in agreement with the streak camera measurements.

**Pulse Fall Time:** In the limit where dephasing by inhomogeneous broadening becomes dominant, $T_2 \ll T_R$, an expression for the radiated intensity can be obtained by retaining all the terms in Eq. (7):

$$I (t) = I (0) e^{-t / \tau_n} + \frac{\hbar \omega_0}{T_R T_2^{\text{eff}}} [g (\omega_0, \Delta \omega_{eff}) + \frac{1}{e^{i \omega_0 / \kappa L T} - 1}] (1 - e^{-t / \tau_n}), \quad (10)$$

where the effective fall time is given by $\tau_n^{-1} = T_2^{-1} (1 + 2 \kappa L - 3 \Lambda^2 e / 4 \pi^2)$. In the absence of propagation loss ($\kappa \simeq 0$), and negligible superradiance rate $1 / T_R$, the fall time of the pulse is dominated by dephasing due to inhomogeneous broadening, $\tau_t = T_2$. An expression for $N$ is similarly found to be

$$N (t) = N (0) e^{-t / T_2} + \frac{\hbar \omega_0}{T_R T_2^{\text{eff}}} [g (\omega_0, \Delta \omega_{eff}) + \frac{1}{e^{i \omega_0 / \kappa L T} - 1}] (1 - e^{-t / T_2}), \quad (11)$$
as $T_1^* \simeq T_2 \ll T_1$. Thus in the dephasing dominated regime both $I(t)$ and $N(t)$ are seen to decay exponentially with a characteristic time of $T_2$ rather than with a sech form which is expected in the regime where dephasing is negligible, $T_2 \gg T_R$. The asymmetry in the pulse shape is naturally anticipated, since $T_D \neq T_2$. The presence of the two timescales and their somewhat independent evolution is clearly manifest in the asymmetry observed in streak camera experiments (See Figs. 3 and 5 of ref. [3], Fig. 4 of ref. [8] along with the expected exponential decay of the pulse.

What determines the dephasing time $T_2$? Normally at high temperatures $T_2$ is determined by Doppler broadening [18]. The fractional Doppler broadening, $\Delta \omega_\ell/\omega = \sqrt{(8 \ln 2) k_B T/M c^2}$, where $M c^2$ is the rest mass energy of the molecule. Defining the inhomogeneous lifetime [18] as $T_2 = 3/\Delta \omega_\ell$, one obtains, for Ar* at $T = 20000 \, \text{K}$, and $\lambda = 400 \, \text{nm}$, $\Delta \omega_\ell = 3.75 \, \text{GHz}$, and a corresponding $T_2 = 130 \, \text{ps}$, comparable to the typical measured fall time in the experiments [3, 8]. Note that $T_2 \propto 1/\sqrt{T}$ and $T_2 \propto \lambda$, denoting that Doppler dephasing time is larger for longer wavelengths, which may explain why the rise time is observed to be almost independent of $\lambda$ whereas the fall time increases linearly with $\lambda$ as observed in Fig. 3 of ref. [7]. But at high pressures, there is a transition from Doppler broadening to collisional processes, which depend on the pressure as well as the collision partner. Correlation experiments described in ref. [8] may be in this regime.

Energy emitted coherently can be obtained by integrating Eq. (10): $I(t) = N h \omega/\pi^2 t^2 e^{-\lambda t}$, the maximum radiated energy is found to be $N h \omega (T_2/T_R)$, indicating that only a fraction of the energy is emitted coherently, only for a time $T_2$ shorter than $T_R$. The rest of the energy is emitted incoherently $\{T_2/T_R = (\alpha_0 L) \ll 1\}$. Thus in the dephasing limited regime one finds that the emitted intensity at first linearly increases with increasing pulsewidth, saturating at a value $T_R$ beyond which $T_2$ becomes irrelevant. The radiated pulsewidth is determined only by $T_R$, as observed in the experiments in ref. [8] (See for instance Fig. 2). $T_R$ can be estimated as $\min\{T_R(\omega_0)\} \approx T_1/N(\omega_0)$. A typical estimate is $T_1/N(\omega_0, \Delta \omega_{\text{eff}}) = (T_1/N)(f_{\text{max}} - f_{\text{min}})T_2$. The total $N$ of $72 \times 10^7$ is obtained at a density of 100 times that of STP for a volume of $(\pi/6) (\lambda = 800 \, \text{nm})^3$. For the visible range, this gives a $T_R$ of 600 ps for $T_1 = 3 \times 10^{-6} \, \text{s}$ for Ar* [3] and an average $T_2$ of 130 ps, consistent with experiments [3].

Emitted Spectrum: The energy emitted at a given $\omega_0$ can be obtained from Eq. (10). Total energy emitted by integrating the energy density $(\pi \alpha_0 L)^{1/2} (g(\omega_0) + e^{h \omega_0/k_B T} - 1)^{-1}$ over the frequency range with the appropriate density of states. At longer wavelengths (or equivalently at higher temperatures) $(e^{h \omega_0/k_B T} - 1)^{-1} \gg g(\omega_0)$, thus the emission spectrum is almost thermal or blackbody-like. This explains why at longer wavelengths (or equivalently in hotter bub-

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