Sonoluminescence as a Cooperative Many Body Phenomenon

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We propose that sonoluminescence occurs due to the cooperative interaction of the matter in the bubble with a radiation field. We illustrate how the collective spontaneous emission of population inverted atomic or molecular states can lead to a time scale of the light pulse which is consistent with that observed in experiments. Pumping by an ultrasound source provides the necessary condition for the inverted states to be correlated over a small volume to trigger sonoluminescence. Experimentally observed role of trace impurities is seen to be consistent in this picture. [S0031-9007(97)04952-1]

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The phenomenon of sonoluminescence (SL), an outburst of a very short light pulse from a gas bubble in a liquid modulated by an ultrasound, has recently generated quite a few interesting experiments [1]. Thermal blackbody or bremsstrahlung radiation [1–3], dynamical Casimir effect [4,5], and collision induced emission [6] among many others [1,2,7,8] have been suggested as possible mechanisms of its origin. In spite of extensive experiments, and equally intriguing theories proposed, the cause of the outburst remains a mystery. In the quantum theories [4,5] first principles calculation of the short time scale (i.e., <10 ps) [9] of the radiated light pulse has not been done, though such a time scale is consistent in this framework. Another important aspect of the experiments [10,11] is that when only a trace of certain solutes (inert gases or other molecules) is present in the liquid solution, SL intensity changes by at least an order of magnitude, and even peaks at such minute concentrations of some of these solutes, though the dielectric constant hardly changes at all. It is not yet clear how to understand this in these approaches [4,5,10]. That the phenomenon of SL is extremely sensitive in its parameter space, and which itself is multidimensional is by now very clear [1,12]. Theoretical interest has also been concerned with the classical hydrodynamic stability and evolution of the bubble [1–3,13]. This classical picture proposes a converging shock wave during the final stages of its contraction, which bounces off the bubble center, compressing the gas to tremendous pressure and temperature, thus causing the outburst. These theories generally are restricted to understanding the bubble dynamics and do not address the *inherent* quantum process of radiation. This paper attempts to address this key issue of the quantum mechanical origin of SL and is complementary to the classical description of bubble dynamics in some of these theories [1–3,13].

We propose a new mechanism to explain the origin of SL as a cooperative many body effect in a two step process. The first step is the population inversion required to obtain a high density of excited atomic or molecular states. As the bubble contracts and reaches its minimum size, gas in the bubble undergoes order(s) of magnitude increase in its pressure and temperature [1,3,13] which makes the gas highly excited or ionized. There results a population inversion of such excited atoms or molecules. This completes the first step. Atoms in the excited states may return to their ground states by the spontaneous emission of radiation. However, if the distance between the excited atoms is smaller than a length scale, say the wavelength of the emitted light, then there could result a phase correlation in the electromagnetic field. The common radiation field of N atoms is just the superposition of the field contributions of the individual component atoms, $\vec{E} = \sum_{i=1}^{N} \vec{E}_i \sim N$, and the intensity of the emitted radiation, $I \sim |\vec{E}|^2$. The total intensity may be written as

$$I \sim \sum_{i,j} \vec{E}_i \vec{E}_j^{*} = \sum_i |\vec{E}_i|^2 + \sum_{i \neq j} \vec{E}_i \cdot \vec{E}_j^{*} \sim N^2. \qquad (1)$$

In case of destructive interference, $I \sim N$, as in normal radiation [14]. If however, the interference term in the last expression is nonvanishing, then $I \sim N^2$. This *cooperative* spontaneous decay is the second step. The collective nature of the emission ensures that the peak intensity of the emitted light goes roughly as N^2. Correspondingly the duration of the outburst for $N \gg 1$ goes as T_1/N, where T_1 is the decay time for a single excited atom [15]. The long-range phase correlation encompassing a large number of component atoms results in the formation of macroscopic quantum coherence (MQC). The MQC state is a superposition of macroscopically distinguishable states and has macroscopic observables. Collective decay contrasts with normal spontaneous decay where individual atoms do not correlate their decay with those in the entire system. Existence of *quantum coherence* is implicit from the phase information contained in the field correlations mentioned above [16]. The creation and spontaneous decay of the collective excited (MQC) state is mediated by the electromagnetic radiation field present in the bubble. One of the factors that may lead to the enhancement of population inversion and MQC is to require metastable states with "a very long lifetime" en route to the excited states.

A complete and thorough calculation of these ideas involves a multiple transition frequency collective decay model [17]. For clarity and to demonstrate the essential physical ideas easily, we follow a much simpler model for the decay of the collective state following the standard superfluorescence model [15] closely. It is not to be construed as a final or complete quantification of these subtle concepts. To treat the problem of coupling of a collection of atoms to a radiation field, we consider the *effective two-level* quantum Hamiltonian operator $\hat{H} = \hat{H}_F + \hat{H}_A + \hat{H}_{AF}$, where $\hat{H}_A = \frac{1}{2}\sum_l \hbar\omega_0\hat{\sigma}_{l3}$ is the atomic part where σ_{li} are the Pauli spin operators for the lth atom, $\hat{H}_F = \sum_{\mathbf{k}\lambda}\hbar\omega_{\mathbf{k}\lambda}\hat{a}^\dagger_{\mathbf{k}}\hat{a}_{\mathbf{k}}$, the electromagnetic field part. Each of the N atoms is considered a two-level prototype system [18]. Thus the lth atom has a ground state $|-\rangle_l$ and an excited state $|+\rangle_l$ to which it may be excited by an external pumping mechanism provided by the contraction of the bubble by the ultrasound in SL. In this simple calculation we assume that all two-level systems have the same transition frequency ω_0. In general the transition frequency ω_{0l} for different atoms is very different and is distributed over a broad spectrum. The dipole interaction term $\hat{H}_{AF}$ is given as

$$H_{AF} = \frac{\omega_0 d}{c}\sum_{\mathbf{k}\lambda l}(g_{\mathbf{k}\lambda l}\hat{a}_{\mathbf{k}} + g^*_{\mathbf{k}\lambda l}\hat{a}^\dagger_{\mathbf{k}})\hat{\sigma}_{l2}\,, \tag{2}$$

where d is the magnitude of the dipole matrix element $\mathbf{d}$ of the two-level system so that $\mathbf{u}d = \mathbf{d}$, and $\mathbf{u} \equiv \mathbf{d}/(\mathbf{d}\cdot\mathbf{d})^{1/2}$. We have thus assumed that all the atoms in the system are identical and have the same dipole matrix element $\mathbf{d}$ [19]. The coupling constant of the atom field interaction in usual notation is $g_{\mathbf{k}\lambda l} = [(2\pi\hbar c^2)/(\omega_{\mathbf{k}\lambda}V)]^{1/2}(\boldsymbol{\epsilon}_{\mathbf{k}\lambda}\cdot\mathbf{u})\exp(i\mathbf{k}\cdot\mathbf{r}_l)$, where $\mathbf{r}_l$ is the position vector of the lth atom. The time evolution of the photon destruction operator is given by

$$\dot{\hat{a}}_{\mathbf{k}}(t) = -i\omega_{\mathbf{k}\lambda}\hat{a}_{\mathbf{k}} - \left(\frac{i\omega_0 d}{c\hbar}\right)\sum_{l=1}^{N} g^*_{\mathbf{k}\lambda l}\hat{\sigma}_{l2}\,. \tag{3}$$

This equation may be formally integrated to obtain

$$\begin{aligned}\hat{a}_{\mathbf{k}}(t) = \hat{a}_{\mathbf{k}}(0)e^{-i\omega_{\mathbf{k}}t} + \frac{i\omega_0 d}{2c\hbar}\sum_{l=1}^{N} g^*_{\mathbf{k}\lambda l}\\ \times[\hat{\sigma}_{l+}(t)\zeta^*(\omega_{\mathbf{k}}+\omega_0)\\ -\hat{\sigma}_{l-}(t)\zeta^*(\omega_{\mathbf{k}}-\omega_0)],\end{aligned} \tag{4}$$

where we define $\hat{\sigma}_{l\pm} \equiv \hat{\sigma}_{l1} \pm i\hat{\sigma}_{l2}$ and $\zeta^*(x) \equiv \frac{P}{x} + i\pi\delta(x)$. We now obtain the rate of the change in the number of photons $\langle\hat{n}_{\mathbf{k}}\rangle = \langle\hat{a}^\dagger_{\mathbf{k}}\hat{a}_{\mathbf{k}}\rangle$ in the field mode $\mathbf{k}$, by using Eqs. (3) and (4) along with their complex conjugates to evaluate $\langle n_{\mathbf{k}}\rangle$ and making the rotating wave approximation (RWA) in the end. The expectation is taken in a state defined as $|\Psi\rangle \equiv |0\rangle|\Phi_N\rangle$ where $|0\rangle$ is the vacuum state of the photon field and $|\Phi_N\rangle$ is an arbitrary N atom state not yet specified. We then have

$$\begin{aligned}\langle\dot{\hat{n}}_{\mathbf{k}}\rangle = \frac{\pi}{2}\left(\frac{\omega_0 d}{c\hbar}\right)^2\sum_{l=1}^{N}\sum_{m=1}^{N} g_{\mathbf{k}\lambda l}g^*_{\mathbf{k}\lambda m}\\ \times\delta(\omega_{\mathbf{k}}-\omega_0)\langle\Phi_N|\hat{\sigma}_{l+}\hat{\sigma}_{m-}|\Phi_N\rangle.\end{aligned} \tag{5}$$

The intensity of the emitted photon pulse at time t in the direction of $\mathbf{k}$ is defined as $I_N(\mathbf{k},t) \equiv \sum_{\omega_{\mathbf{k}}}\hbar\omega_{\mathbf{k}}\langle\dot{\hat{n}}_{\mathbf{k}}\rangle$. All atoms being identical the expectation values in Eq. (5) depend on just two indices, say a and b, used to label the atoms. The sum over $\omega_{\mathbf{k}}$ is converted into an integral in the continuum limit. Thus using Eq. (5) we get

$$\begin{aligned}I_N(\mathbf{k},t) = \frac{I_1(\mathbf{k},0)N}{4}\{2(1+\langle\Phi_N|\hat{\sigma}_{a3}|\Phi_N\rangle)\\ +[N\Gamma(\mathbf{k})-1]\\ \times\langle\Phi_N|\hat{\sigma}_{a+}\hat{\sigma}_{b-}|\Phi_N\rangle\},\end{aligned} \tag{6}$$

where we define $I_1(\mathbf{k},0) \equiv (3\hbar\omega_0\sum_\lambda|\boldsymbol{\epsilon}_{\mathbf{k}}\cdot\mathbf{u}|^2)/8\pi T_1$, $1/T_1 \equiv 4\omega_0^3 d^2/3c^3\hbar$, and the structure factor $\Gamma(\mathbf{k}) \equiv |\sum_{l=1}^{N}[\exp(i\mathbf{k}\cdot\mathbf{r}_l)]/N|^2$. The many atom state $|\Phi_N\rangle$ in general is an MQC state. For the present simple calculation we choose

$$\begin{aligned}|\Phi_N\rangle \equiv \prod_{n=1}^{N}|\theta,\phi\rangle_n \quad\text{where}\\ |\phi,\theta\rangle_n \equiv \sin(\theta/2)e^{-i\phi/2}|+\rangle_n + \cos(\theta/2)e^{i\phi/2}|-\rangle_n\,.\end{aligned} \tag{7}$$

The dimensionless energy of the N atom system is given by $W_N \equiv (1/2)\langle\Phi_N|\hat{\sigma}_{n3}|\Phi_N\rangle$ which simplifies with the use of Eq. (7) to $W_N(t) = (-1/2)N\cos[\theta(t)]$. So far we have ignored the effect of inhomogeneous broadening which may be incorporated into Eq. (6) by multiplying $\Gamma(\mathbf{k})$ by a factor $H(t) = \exp(-|t|/T_2^*)$. Using these results in Eq. (6) we get the emitted photon intensity per unit solid angle around the direction of $\mathbf{k}$ at time t as

$$\begin{aligned}I_N(\mathbf{k},t) = [I_1(\mathbf{k},0)/(4N)][N+2W_N(t)]\\ \times\{2N+[N-2W_N(t)][NH(t)\Gamma(\mathbf{k})-1]\}.\end{aligned} \tag{8}$$

Conservation of energy implies that the energy lost by the atoms should be gained by the photon field. This can be derived explicitly in the RWA and hence we get $-\hbar\omega_0\dot{W}_N(t) = I_N(t)$, where $I_N(t) \equiv \oint I_N(\mathbf{k},t)d\Omega_{\mathbf{k}}$. The integral is over all directions of $\mathbf{k}$. This gives the result

$$\begin{aligned}-\dot{W}_N(t) = I_N(t)/(\hbar\omega_0)\\ = [1/(4T_1)][N+2W_N(t)]\\ \times[N\mu - 2\mu W_N(t) + 2].\end{aligned} \tag{9}$$

We choose the initial state as one that has all atoms in their respective excited state $|+\rangle$ (i.e., complete inversion), which implies $W(0) = N/2$. We may now solve Eq. (9) for $W_N(t)$ and $I_N(t)$ in the limit $H(t) \approx 1$ to get [20]

$$W_N(t) = \frac{1-(1+N\mu)\tanh[(t-t_0)/2t_p]}{2\mu} \tag{10}$$

and

$$I_N(t) = \frac{\hbar\omega_0(1+N\mu)^2\,\mathrm{sech}^2[(t-t_0)/2t_p]}{4\mu T_1}, \tag{11}$$

where

$$\mu \equiv -N^{-1} + \oint [I_1(\mathbf{k},0)/I_0]\Gamma(\mathbf{k})H(t)\,d\Omega_{\mathbf{k}}\,, \qquad (12)$$

$I_0 \equiv \oint I_1(\mathbf{k},0)\,d\Omega_{\mathbf{k}}$, $t_0 \equiv t_p \ln(N\mu)$, and $t_p \equiv T_1/(1 + N\mu)$.

The solution for $W_N(t)$ from Eq. (11) can be used in Eq. (8) to get the explicit temporal and directional dependence of the emitted intensity. Equation (11) is a key result which we shall now discuss. Let us assume that the volume of the emitting atoms is a perfect sphere of radius R. Converting sums into integrals in the continuum limit while evaluating μ, it may be shown that $N\mu = 9(N-1)\,(kR)^{-6}[kR\cos(kR) - \sin(kR)]^2$, where $k \equiv |\mathbf{k}|$. As expected the radiation in this limit is radially symmetric.

To get physically relevant numbers we choose $k = 2\pi/(400\text{ nm})$, $T_1 = 10$ ns, and the density to be about 100 times that of the gas at STP (i.e., 100 times Avagadro's number in 0.0224 m^3; note that the actual density may have a radial dependence but the estimates of the intensity and duration of the SL pulse will remain qualitatively unchanged), $R = 50$ nm. This gives a total number of radiating atomic or molecular entities of about 1.4×10^6. This would be measured as the total number of photons emitted which is in the ballpark of the measured value [1]. Then using definitions in Eqs. (12) we get $t_p = 8$ fs and $t_0 \approx 0.1$ ps and the total energy emitted per pulse of about 4 MeV. We now note that the measured SL spectrum [10] is a continuum or at least has a large distribution in wavelength from about 200 to 700 nm. Obviously, the million or so atoms that produce this spectrum are being excited to various different levels; i.e., the lth atom has a transition frequency ω_{0l} and they are not all equal as assumed here. Equation (11) is derived in a single transition frequency (ω_0) model and hence does not incorporate the different levels of excitation. In a generalization to a multiple transition frequency model with various excitation levels, the essential features such as the short time scale and large peak intensity of Eq. (11) will be retained and the distribution of various excitation levels deduced [17]. The spectrum would depend on this distribution of excited states and hence the preparation of the initial MQC state and its dephasing is very important. The initial MQC state being driven by the thermal or quantum fluctuations, the characteristics of such a driving field would be reflected in the spectrum [17]. If other incoherent interactions are neglected, then the various types of two-level states will be occupied typically in a Planck distribution. Nonetheless, the enhanced distribution corresponding to resonance lines could appear as resonance peaks on top of this continuum spectrum [21]. Besides, though the precise trigger for the radiation process to begin is provided by the coupling of the MQC state to quantum fluctuations, other noncooperative processes may increase the pulse width t_p [20]. In this simple single transition frequency model considered here the time scale of the pulse width $t_p \sim 10$ fs is consistent with the experimental upperbound and the total emitted energy 4 MeV is in the right order of magnitude [1,9]. In the general multiple transition frequency model this pulse width t_p will be somewhat increased.

The emitted light is expected to be coherent and it should be possible to test for this property. An investigation into the spatiotemporal coherence of the SL source, its spatial size and pulse width using photon correlations such as the Hanbury-Brown-Twiss effect [22,23] or other techniques, will provide one of the tests of our proposed SL mechanism. Some other previously proposed mechanisms [1–3,6,7] for SL would entail a chaotic emitted light which would show the following properties in a photon counting experiment performed for any of its filtered single modes [24]: (i) The photons would be bunched and the degree of second order coherence $g^{(2)}(0) = 2$; (ii) the probability of finding m photons in a given measurement which gives an average of $\langle m\rangle$ photons would be $P_m = \langle m\rangle^m(1 + \langle m\rangle)^{-1-m}$.

If an MQC exists during anytime in the SL process then at least some of the emitted light would show a coherent character. However, for purely coherent filtered single mode light [24] (i) the photons would be antibunched and $g^{(2)}(0) = 1$, and (ii) P_m would be a Poissonian distribution. We note that along with the coherent emission proposed in MQC there will also be some chaotic radiation because of (a) incoherent preparation of the initial MQC state from a thermal (blackbody) field, and (b) there may be chaotic radiations from other noncooperative processes. Hence, purely coherent light may not be seen in experiment [instead of $g^{(2)}(0) = 1$, we expect $1 \le g^{(2)}(0) < 2$, presumably close to 1].

There are three possible ways one may obtain a directional dependence in the emitted light: (i) Nonsphericity in the bubble shape may cause the emitted light to undergo refraction or diffraction as may be the case in Ref. [22] and (ii) nonsphericity in the MQC source (whose size could be smaller than the already-compressed bubble size). We include this via μ in the calculation. These first two types of nonuniformities will probably have a long relaxation time scale. (iii) An inherent quantum directionality could be present in each SL light pulse, which would be random from pulse to pulse. Unlike the case in superfluorescence experiments we do not have an incident coherent light causing the inversion of atomic states. We therefore do not expect any large dipole radiation of this third type. Though each *individual single bubble SL pulse* will have some of the above coherent character analyzing averages over many pulses needs caution.

The importance of the thermal conductivity of dissolved gases, their polytropic index, viscosity of the solution, etc. have already been discussed earlier [1,2,21,25]. Hiller *et al.* [10] have also mentioned the possible role of

Penning ionization. We emphasize three criteria which in combination could enhance SL in our picture, namely, that the dissolved gases or solutes (1) have long-lived metastable states, which (2) are high enough in energy to be able to excite or ionize other atoms of their own or other types, and (3) that these states efficiently transfer their energy to other atoms and molecules in inelastic collisions. The rare gases certainly satisfy these conditions. Even trace amounts of such impurities may enhance the inversion of states by an order of magnitude by chain stepwise excitations as occurs in plasmas. The observed peak in SL emission at *precise* concentrations of noble gases [10,26] may then support such an explanation. The reverse effect of quenching of excited states by trace impurities may lead to a dramatic drop in SL intensity, as has also been observed [11,26]. From Eq. (11) the total SL intensity should scale as the square of the dipole moment of the emitting atom or molecule. The increase in SL intensity with the dipole moment of the solution molecules (which may be present in the bubble as well) has also been recorded qualitatively [25].

We have discussed a collective, quantum atomistic idea for the origin of SL. Plausible values of input parameters give estimates of the emitted energy per burst and the short time scale of the SL pulse consistent with experiment. Experimentally observed critical dependence of SL on trace impurities may be understood in this picture. Study of the emitted photon bursts [22,23] would reveal the volume involved in the radiation process, the time duration of the SL bursts, the two photon correlations, and other coherence properties which would serve as adequate tests of the proposed mechanism. The validity of the proposed mechanism will be explored in greater detail elsewhere [17].

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[18] Here as well as later the term "atom" is used synonymously with any atomic or molecular system which may *effectively be modeled* as a two-level system interacting with the electromagnetic field with a dipole interaction. For instance, the excited state could be an electronic excitation of a single atom or a vibrational or rotational excitation of an ionic molecule, etc.

[19] We ignore the level degeneracies of the atomic system, doppler broadening, loss, and any dephasing mechanism other than spontaneous emission.

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