Quantum Optical Heating in Sonoluminescence Experiments

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Abstract. Sonoluminescence occurs when tiny bubbles filled with noble gas atoms are driven by a sound wave. Each cycle of the driving field is accompanied by a collapse phase in which the bubble radius decreases rapidly until a short but very strong light flash is emitted. The spectrum of the light corresponds to very high temperatures and hints at the presence of a hot plasma core. While everyone accepts that the effect is real, the main energy focussing mechanism is highly controversial. Here we suggest that the heating of the bubble might be due to a weak but highly inhomogeneous electric field as it occurs during rapid bubble deformations [A. Kurcz et al. (submitted)]. It is shown that such a field couples the quantised motion of the atoms to their electronic states, thereby resulting in very high heating rates.

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INTRODUCTION

Sonoluminescence is a phenomenon that derives from the acoustic cavitation of noble gas atoms [1]. There are two classes of sonoluminescence: multi-bubble [2, 3] and single-bubble sonoluminescence [4, 5]. Single bubble sonoluminescence is characterized by the emission of a strong light flash over a very short period of time from a single extremely hot gas bubble. Under appropriate conditions, the acoustic force on a bubble can balance against its buoyancy, holding a bubble stable in the liquid by acoustic levitation. Such a bubble is typically quite small compared to an acoustic wavelength and is capable to confine the particles of the trapped van der Waals gas close to their covolume. For specialized conditions, a single, stable, oscillating gas bubble can be forced into such large amplitude pulsations that it produces sonoluminescence during each and every acoustic cycle.

A typical single-bubble sonoluminescence cycle is shown in Fig. 1(a). Most of the cycle, the bubble behaves isothermal (c.f. Fig. 1(b)). Point A marks the beginning of the collapse phase in which the bubble approaches its minimum radius of about 0.5 µm very rapidly with supersonic speed. Here, the bubble becomes thermically isolated from the surrounding liquid. In Point B, the temperature within the bubble significantly increases with a heating rate of $10^{10} - 10^{11}$ K/s and a strong light flash emerges which last for about 40ps. Point C denotes the beginning of the expansion phase in which the bubble oscillates around its equilibrium radius until it regains stability.

The emitted light mainly consist of a continuum of blackbody or Bremsstrahlung radiation. Detailed measurements of the light spectra indicate temperatures above $10^4$ K [6, 7, 8]. It is even possible to observe light emission in the ultraviolet regime which hints
at temperatures of about $10^6$ K in a bubble driven at $1$ Mhz [9]. Emission lines from transitions between high energy states of noble gas atoms which cannot be populated thermally [10, 11] point at the formation of an opaque plasma core [13]. Evidence for a plasma core has also been found in multi-bubble sonoluminescence experiments [12].

The time dependence of the bubble radius and its nearly adiabatic compression are theoretically well understood up to a certain point when it approaches the minimum radius [5, 13]. What is the state of the bubble during the last part of the collapse phase and the conditions that lead to these enormous heating rates up to very high temperatures is still controversial. Here we summarise an idea which suggests that the heating is due to the presence of a highly inhomogenous electric field as it occurs during rapid bubble deformations [14]. This field couples the motion of the noble atoms to their electronic degrees of freedom. When combined with spontaneous emission from the atoms, a quantum optical heating process can occur. Similar couplings are responsible for the cooling of ions in ion trap experiments [15].

**THE BASIC IDEA**

Approaching its minimum radius close to point $B$ in Fig. 1(a), the bubble is no longer in a thermal equilibrium. Suddenly, an increase in entropy occurs which is based on highly irreversible processes. It causes a temperature increase much higher than what can be caused by thermodynamic heating processes (c.f. Fig. 1(b)). In the following we address two questions: Why does the bubbles need to be filled with noble gas atoms? What is the main energy focussing mechanism during the collapse phase of the bubble?

Close to point $B$, the mean distance between the noble gas atoms becomes so small that interactions between them can be described by a Lennard-Jones potential. Indeed, the physical condition of the bubble becomes that of a solid state system. The atoms experience an equilibrium between repulsive interatomic forces due to overlapping orbitals and attractive forces due to the van der Waals interaction. Thus, any significant gain in temperature has to be caused by vibrational motion driven into the quantum regime. Furthermore, the presence of light requires the assumption of an open quantum
system.

To model the resulting strong confinement of the atoms, we place each of them into an approximately harmonic trapping potential. This allows us to quantise the atomic motion during the collapse phase, just before the maximum compression of the bubble. Around this point, the motional states of each atom can be described by phonons with frequency $\nu$. In the next section, we show that the gradient of an electric field inside the bubble establishes a coupling between the electronic and the quantised motional states of each noble gas atom. The origin of the field can be explained by an inhomogeneous charge distribution of ionized species from the dissolved liquid due to rapid bubble deformations.

For simplicity we assume that the atoms are effective two-level systems with ground state $|0\rangle$ and excited state $|1\rangle$. The corresponding interaction Hamiltonian contains terms that result in the excitation and de-excitation of each atom accompanied by the creation and the annihilation of a phonon. Also crucial is the presence of a large spontaneous decay rate $\Gamma$ of the excited state $|1\rangle$ which keeps the atoms predominantly in their ground state. Although these processes are highly non-resonant, they result in a significant change of the mean phonon number per atom and increase the temperature inside the bubble by many orders of magnitude, even within a few nanoseconds.

Suppose an atom is initially in its ground state and possesses exactly $m$ phonons, as shown in Fig. 2a. We denote this state by $|0, m\rangle$. Notice that phonons are bosons which are described by annihilation operators $b$ with $[b, b^\dagger] = 1$. Consequently, a transition into the state $|1, m+1\rangle$ occurs with a rate proportional to $\sqrt{m+1}$, while the rate for a transition into the state $|1, m-1\rangle$ scales only as $\sqrt{m}$. Since the spontaneous decay rate of the atom is relatively large, such a transition is immediately followed by an irreversible and predominantly non-radiative transition back into $|0\rangle$. This transfers the atom either into its initial state $|0, m\rangle$ or into the states $|0, m-1\rangle$ and $|0, m+1\rangle$, respectively. The net effect is an increase of the mean phonon number per atom, i.e. heating, since the phonon population in the latter state is higher than the phonon population in $|0, m-1\rangle$.

THE TIME EVOLUTION OF THE SYSTEM

We now consider a single noble gas atom at the position $r$. This atom is typical for the many atoms inside the bubble. Its dipole Hamiltonian equals

$$H_{\text{int}} = e\mathbf{D} \cdot \mathbf{E}(r)$$

with $e$ being the charge of a single electron, the (real) atomic dipole moment

$$\mathbf{D} = D_{01} \sigma^- + \text{H.c.}$$

$\sigma^+ \equiv |1\rangle \langle 0|$, $\sigma^- \equiv |0\rangle \langle 1|$, and where $\mathbf{E}$ is the electric field inside the bubble. For simplicity, we assume that all field components point in the direction of a single unit vector $\hat{k}$. This allows us to write $\mathbf{E}(r)$ as

$$\mathbf{E}(r) = \sum_k E_k e^{i\hat{k} \cdot r} + \text{c.c.}$$
FIGURE 2. (a): Level configuration of a single atom-phonon system indicating the immediately relevant transitions, if the atom is initially in $|0, m\rangle$. $\Omega$ and $\Lambda$ denote coupling constants and $\Gamma$ is the spontaneous decay rate of level 1. (b): The mean phonon number $m$ as a function of time for $\nu = 10$ MHz while $\Omega = 10^6$ Hz, $\Lambda = 10^{12}$ Hz, $\Gamma = 10^{13}$ Hz, and $\omega_0 = 10^{15}$ Hz. Good agreement is found between the numerical solution of the full rate equations (9) and (10) and Eq. (13) (shaded area).

with amplitudes $E_k$ and wave vectors $k = k \hat{k}$. Moreover, we consider the atomic motion in the $\hat{k}$-direction as quantised with $b$ being the corresponding phonon annihilation operator. Then $\hat{k} \cdot (r - R) = \Delta x (b + b^\dagger)$. Here $R$ is the current equilibrium position of the noble gas atom with mass $M$ and $\Delta x = \sqrt{\hbar/2MV}$ is the width of its ground state wave function in the respective vibrational mode. If the atom is well localized within the wavelength of its trapping potential, the Lamb-Dicke approximation allows us to assume that $\exp(i \hat{k} \cdot (r - R)) = 1 + i \Delta x (b + b^\dagger)$ [15, 16]. Substituting this into Eq. (1), we obtain the interaction Hamiltonian

$$H_{\text{int}} = \hbar \Omega (\sigma^- + \sigma^+) + \hbar \Lambda (b + b^\dagger)(\sigma^- + \sigma^+)$$

with the (real and positive) coupling constants

$$\Omega \equiv \left(2e/\hbar\right) \sum_k D_{01} \cdot \text{Re} \left(E_k e^{ik \hat{k} \cdot R}\right),$$

$$\Lambda \equiv -\left(2e \Delta x/\hbar\right) \sum_k k D_{01} \cdot \text{Im} \left(E_k e^{ik \hat{k} \cdot R}\right).$$

This Hamiltonian is essentially a Jaynes-Cummings Hamiltonian with $\Lambda$ being proportional to the gradient of $\Omega$ in the direction of the quantised motion of the atom, i.e. $\Lambda = \Delta x \hat{k} \cdot \nabla \Omega(R)$. A strong atom-phonon coupling therefore does not necessarily require the presence of a strong electric field. It only requires a highly inhomogeneous field inside the bubble. In the following, we neglect interactions between the noble gas atoms other than the ones already included in the harmonic trapping potential of each particle. Dissipation in form of spontaneous photon emission from the atomic state $|1\rangle$ is taken into account by the master equation [16]

$$\dot{\rho} = -\frac{i}{\hbar} \left[H_{\text{int}} + \hbar \omega_0 (\sigma^+ \sigma^- + \hbar \nu b^\dagger b), \rho\right] + \Gamma \left[\sigma^- \rho \sigma^+ - \frac{1}{2} \sigma^+ \sigma^- \rho - \frac{1}{2} \rho \sigma^+ \sigma^- \right].$$

(6)
Here \( \hbar \omega_0 \) and \( \hbar \nu \) are the energy of the atomic state \( |1\rangle \) and of a single phonon.

Eq. (6) can now be used to obtain a closed set of rate equations. Its major quantities are the phonon number \( m \equiv \langle b^\dagger b \rangle \) and

\[
X_{1,2} \equiv \langle \sigma_1 \rangle, \quad X_3 \equiv \langle \sigma^+ \sigma^- - \sigma^- \sigma^+ \rangle, \quad Y_1 \equiv \langle b + b^\dagger \rangle, \quad Y_2 \equiv \langle i(b - b^\dagger) \rangle, \\
Y_3 \equiv \langle b^2 + b^\dagger 2 \rangle, \quad Y_4 \equiv \langle i(b^2 - b^\dagger 2) \rangle, \quad Z_{1,2} \equiv \langle \sigma_1,2(b + b^\dagger) \rangle, \quad Z_{3,4} \equiv \langle i(\sigma_1,2(b - b^\dagger)) \rangle
\]  

(7)

with the Pauli operators \( \sigma_i \equiv \sigma^+ + \sigma^- \) and \( \sigma_2 \equiv i(\sigma^- - \sigma^+) \). Here we assume

\[
\omega_0 \gg \nu, \Gamma, \Omega, \Lambda \quad \text{and} \quad m \gg 1
\]

and approximate the expectation value of operators of the form \( \langle B \sigma_3 \rangle \) by \( \langle B \rangle \langle \sigma_3 \rangle \). The latter applies when the expectation value of \( B \) is about the same for an atom in \( |0\rangle \) and for an atom in \( |1\rangle \). Eq. (6) then yields

\[
\dot{m} = \Lambda Z_3, \quad \dot{X}_3 = 2(\Omega X_2 + \Lambda Z_2) - \Gamma (X_3 + 1), \quad \dot{Y}_1 = -\nu Y_2, \\
\dot{Y}_2 = 2\Lambda X_1 + \nu Y_1, \quad \dot{Y}_3 = -2(\nu Y_4 + \Lambda Z_3), \quad \dot{Y}_4 = 2(\nu Y_3 + \Lambda Z_1),
\]

(9)

and

\[
\dot{X}_1 = -\omega_0 X_2, \quad \dot{X}_2 = -2(\Omega + \Lambda Y_1) X_3 + \omega_0 X_1, \quad \dot{Z}_1 = -\omega_0 Z_2, \quad \dot{Z}_3 = 2\Lambda - \omega_0 Z_4, \\
\dot{Z}_2 = -2(\Omega Y_1 + \Lambda Y_3 + 2\nu m) X_3 + \omega_0 Z_1, \quad \dot{Z}_4 = -2(\Omega Y_2 + \Lambda Y_4) X_3 + \omega_0 Z_3
\]

(10)

up to first order in \( 1/\omega_0 \). In the beginning of each sonoluminescence cycle the particles experience neither a strong trapping potential nor the presence of an inhomogeneous electric field inside the bubble. We can therefore assume that the coherences defined in Eq. (7) are initially zero and that the atom is in its ground state. Condition (8) allows us to simplify the above rate equations via an adiabatic elimination of Eq. (10). Doing so we obtain a set of equations where the derivatives of \( X_3, Y_1, \) and \( Y_2 \) decouple from the rest. Solving them for the case of a relatively strong atom-phonon coupling constant \( \Lambda \) with \( \Lambda \gg \Omega \) and \( 4\Lambda^2 > \nu \omega_0 \) yields

\[
X_3(t) = -1, \quad Y_1(t) = \frac{4\nu \Omega \Lambda}{\lambda^2 \omega_0} \cdot \left[ \cosh(\lambda t) - 1 \right], \quad Y_2(t) = -\frac{4\Omega \Lambda}{\lambda \omega_0} \cdot \sinh(\lambda t)
\]

(11)

with \( \lambda \equiv \nu \left( 4\Lambda^2 / \nu \omega_0 - 1 \right)^{1/2} \) up to first order in \( 1/\omega_0 \). For times \( t \) of the order of \( 1/\lambda \), \( Y_1 \) and \( Y_2 \) are of the order of \( 1/\omega_0 \). Taking this into account, we find that \( Z_1 = -2\Lambda(2m + Y_3) / \omega_0 \) and \( Z_3 = -2\Lambda Y_4 / \omega_0 \) in first order in \( 1/\omega_0 \). The variables \( m, Y_3, \) and \( Y_4 \) in Eq. (9) hence evolve according to

\[
\dot{m} = -\frac{2\Lambda^2}{\omega_0} Y_4, \quad \dot{Y}_3 = \frac{2(2\Lambda^2 - \nu \omega_0)}{\omega_0} Y_4, \quad \dot{Y}_4 = -\frac{8\Lambda^2}{\omega_0} m - \frac{2(2\Lambda^2 - \nu \omega_0)}{\omega_0} Y_3.
\]

(12)

For \( m(0) = m_0 \) and \( Y_3(0) = Y_4(0) = 0 \), this yields

\[
m(t) = m_0 + \frac{8\Lambda^4}{\lambda^2 \omega_0^2} m_0 \sinh^2(\lambda t). \]

(13)
As one can see in Fig. 2(b), Eq. (13) describes an approximately exponential heating process as long as a relatively large decay $\Gamma$ secures that the atom remains in the ground state predominantly $^1$. Taking into account typical experimental parameters, the phonon energy in the bubble can easily increase by a factor ten or more, even within a few nanoseconds. Using the relation $m \cdot h\nu = k_B T$, our model can easily predict temperatures well above $10^4$ K inside the bubble.

CONCLUSION

We attribute the sudden concentration of energy in sonoluminescence experiments to the heating of strongly confined noble gas atoms by a highly inhomogeneous electric field. The time evolution of each atom is dominated by non-energy conserving processes, which result in a permanent increase of its mean phonon number $m$ when combined with spontaneous emission. Our model does not contradict current models for the description of sonoluminescence experiments, but explains previously controversial aspects of this phenomenon. It is based on a quantum optical approach that is routinely used to describe the laser cooling of tightly trapped ions $^{15}$.

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$^1$ The assumption of a relatively high spontaneous decay rate can be justified by the presence of collective effects inside the van der Waals gas formed by the noble gas atoms.