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# Temperature of a Compressed Bubble with Application to Sonoluminescence

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The rise in temperature from the adiabatic compression of a bubble is computed in thermodynamic mean field (van der Waals) theory. It is shown that the temperature rise is higher for the noble gas atoms than for more complex gas molecules. The adiabatic temperature rise is shown to be sufficient for producing sonoluminescence via the excited electronic states of the atoms.

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## I. INTRODUCTION

There has been considerable interest in the dynamics of single bubble sonoluminescence[1, 2, 3, 4]. It is found that noble gases (within the bubble) greatly enhance the emitted light intensity when compared to bubbles filled with other gases [5, 6, 7, 8, 9, 10, 11]. The theoretical interpretation of this experimental fact will be discussed in the work which follows.

The fluid mechanics of bubble motion[12, 13, 14] is thought to play a central role in the dynamics of sonoluminescence. The starting point of theoretical bubble dynamics has been the Rayleigh-Plesset equation [15, 16, 17]. An important feature of the experimental bubble fluid dynamics is the exceedingly high temperatures[18, 19, 20] that can be achieved by merely decreasing the radius of a bubble. This is especially true when noble gas atoms are contained within the bubble [21, 22, 23, 24, 25, 26]. Although the dynamics of bubble compression appears to be intimately connected to the high noise temperature of emitted light pulses, the process of light emission has not yet been firmly established [27, 28, 29, 30, 31].

Our purpose is to calculate the maximum temperature of the contents inside the bubble on the basis of fluid mechanics[32, 33, 34] using a mean field (van der Waals) thermodynamic equation of state for adiabatic compression. We find that the resulting temperatures are sufficiently high to produce light from sonoluminescence if not from mere incandescence. We also estimate the concentration of atoms with excited electronic states which arise from an adiabatic compression of the fluid within the bubble.

In Sec.II, we derive the thermodynamic identities required to compute the temperature rise during adiabatic compression of a bubble. The mean field (van der Waals) thermodynamic equation of state is discussed in Sec.III wherein numerical results are presented for the case of argon. Arguments are presented which explain why noble gas atoms are particularly suited for achieving high temperatures for small bubble radii. In Sec.IV, a Lagrangian formulation of the bubble dynamics will be employed to discuss in more detail the role of adiabatic processes. The internal chemistry[35] of the bubble will be discussed in Sec.V, while the physical aspects of the chemical reaction

will be further explored in the concluding Sec.VI.

## II. THERMODYNAMICS

If  $\epsilon(s, v)$  is the energy per particle of a fluid *inside the bubble* as a function of volume per particle  $v$  and entropy per particle  $s$ , then

$$d\epsilon = Tds - pdv, \quad (1)$$

where  $T$  and  $p$  denote (respectively) the temperature and pressure of the fluid. Alternatively, one may employ the free energy per particle

$$f(T, v) = \inf_{\epsilon} \{ \epsilon - Ts(\epsilon, v) \}, \quad (2)$$

which obeys

$$df = -sdT - pdv. \quad (3)$$

Condensed matter often (but not always) exhibits a lowering of the temperature when adiabatically expanded and a temperature rise when adiabatically compressed. The thermal coefficient describing such a process may be defined as

$$\eta = - \left( \frac{v}{T} \right) \left( \frac{\partial T}{\partial v} \right)_s. \quad (4)$$

From Eq.(1) it follows that

$$- \left( \frac{\partial T}{\partial v} \right)_s = \left( \frac{\partial p}{\partial s} \right)_v = \left( \frac{\partial p}{\partial T} \right)_v \left( \frac{\partial T}{\partial s} \right)_v. \quad (5)$$

Eqs.(4) and (5) imply

$$\eta = \frac{v}{c_v} \left( \frac{\partial p}{\partial T} \right)_v, \quad (6)$$

where

$$c_v = T \left( \frac{\partial s}{\partial T} \right)_v \quad (7)$$

is the specific heat at constant volume. Similarly

$$c_p = T \left( \frac{\partial s}{\partial T} \right)_p \quad (8)$$

is the specific heat at constant pressure which obeys

$$c_p = c_v + T \left( \frac{\partial v}{\partial T} \right)_p \left( \frac{\partial p}{\partial T} \right)_v. \quad (9)$$

On the one hand, we have from Eqs.(4), (5) and (7) that

$$- \left( \frac{\partial T}{\partial v} \right)_s = \frac{T}{c_v} \left( \frac{\partial p}{\partial T} \right)_v. \quad (10)$$

On the other hand, with

$$\gamma = \left( \frac{c_p}{c_v} \right) \quad \text{and} \quad \beta = \frac{1}{v} \left( \frac{\partial v}{\partial T} \right)_p \quad (11)$$

Eqs.(9), (10) and (11) imply

$$- \left( \frac{\partial T}{\partial v} \right)_s = \left( \frac{\gamma - 1}{v\beta} \right). \quad (12)$$

The temperature rise corresponding to an adiabatic compression is then calculated from

$$\ln \left( \frac{T_f}{T_i} \right) = - \int_{v_f}^{v_i} \frac{1}{T} \left( \frac{\partial T}{\partial v} \right)_s dv, \quad (13)$$

where  $(\partial T / \partial v)_s$  may be calculated from either Eq.(10) or Eq.(12).

For the case of an ideal gas with  $P_0 v = k_B T$ ,  $\gamma = \text{const}$  and  $T\beta_0 = 1$ , Eqs.(12) and (13) imply

$$\left( \frac{T_f}{T_i} \right)_{(\text{ideal gas})} = \left( \frac{v_i}{v_f} \right)^{(\gamma-1)} = \left( \frac{R_i}{R_f} \right)^{3(\gamma-1)}, \quad (14)$$

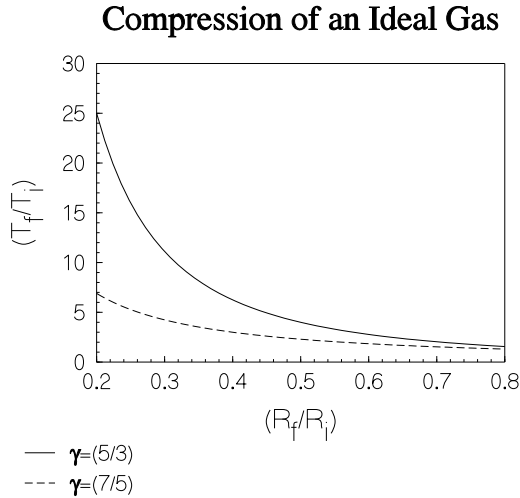


FIG. 1: For the adiabatic compression of an ideal gas in a bubble from radius  $R_i \rightarrow R_f < R_i$ , the temperature rise is described by  $T_i \rightarrow T_f > T_i$ . For the case of noble gas atoms ( $\gamma = 5/3$ ) the temperature rise is much more steep than the case of tumbling diatomic molecules ( $\gamma = 7/5$ ).

where  $N$  particles in a bubble of radius  $R$  are described by the volume

$$V = Nv = \left( \frac{4\pi R^3}{3} \right). \quad (15)$$

The adiabatic compression of an ideal gas is plotted in FIG.1. For noble gas atoms ( $\gamma = 5/3$ ) the rise in temperature is substantial [36]. If the radius drops by a factor of five, the ideal gas can exhibit a temperature rise from (say) room temperature to a temperature  $T \sim 7 \times 10^3 \text{ K}$  [38, 39, 40, 41, 42, 43] which is approximately the surface temperature of the sun. For an ideal gas of tumbling diatomic molecules ( $\gamma = 7/5$ ) the rise in temperature upon adiabatic compression is much less steep.

The general situation follows from the thermodynamic Eqs.(10) and (13). For a complicated molecule, the internal degrees of freedom of the constituent molecules give rise to a high specific heat  $c_v$ . The adiabatic compression of a fluid with a high heat capacity yields an only moderate rise in temperature. On the other hand, the noble gas atoms have at most internal electronic degrees of freedom and these lie dormant for reasonably low temperatures. Thus, noble gas atom fluids have a low heat capacity and a resulting high temperature rise on an adiabatic compression. These qualitative considerations give more than just a little insight into why noble gas atoms (in particular Ar atoms) have played such an important role in laboratory experiments on sonoluminescence.

The sonoluminescent bubble can be so compressed that the fluid contained within the bubble is far from ideal. However, the above thermodynamic considerations are general and can be applied to fluids which are far from being ideal gases. In the mean field (van der Waals) theory of argon fluids, the equations of state can be worked out in detail.

### III. VAN DER WAALS THEORY

The free energy per particle of an ideal gas of noble gas atoms may be written as

$$f_0(T, v) = -k_B T \ln \left( \frac{v}{\lambda_T^3} \right), \quad (16)$$

where the thermal wavelength of an atom of mass  $m$  is given by

$$\lambda_T = \sqrt{\frac{2\pi\hbar^2}{mk_B T}}. \quad (17)$$

In mean field theory, the hard core of the two atom potential is included by the “free volume” replacement  $v \rightarrow v - b$  while the attractive part of the potential is simulated by subtracting (from the free energy) an attractive mean potential term ( $a/v$ ) proportional to the

fluid density. Thus, the “mean field” theory modification to the ideal gas Eq.(16) is given by

$$\bar{f}(T, v) = -k_B T \ln \left( \frac{v-b}{\lambda_T^3} \right) - \left( \frac{a}{v} \right). \quad (18)$$

The mean field theory pressure  $\bar{p} = -(\partial \bar{f} / \partial v)_T$  which follows from Eq.(18) is that of van der Waals

$$\bar{p} = \left( \frac{k_B T}{v-b} \right) - \left( \frac{a}{v^2} \right), \quad (19)$$

while the specific heat at constant volume follows from Eqs.(17) and (18) as

$$\bar{c}_v = T \left( \frac{\partial \bar{s}}{\partial T} \right)_v = -T \left( \frac{\partial^2 \bar{f}}{\partial T^2} \right)_v = \left( \frac{3k_B}{2} \right). \quad (20)$$

Eqs.(10), (19) and (20) imply that

$$- \left( \frac{\partial T}{\partial v} \right)_s = \left( \frac{2T}{3(v-b)} \right) \quad (\text{van der Waals}). \quad (21)$$

Finally, Eqs.(13) and (21) yield the mean field theory adiabatic temperature rise in a noble gas atom fluid upon adiabatic compression; It is

$$\begin{aligned} \left( \frac{T_f}{T_i} \right)_{(\text{van der Waals})} &= \left( \frac{v_i - b}{v_f - b} \right)^{2/3} \\ &= \left( \frac{1 - y^3}{(R_f/R_i)^3 - y^3} \right)^{2/3}, \quad (22) \end{aligned}$$

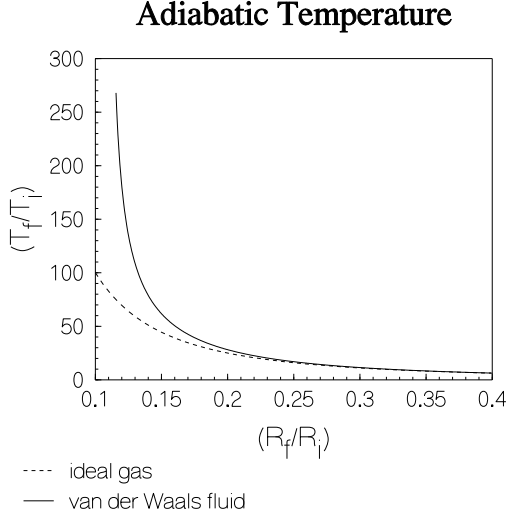


FIG. 2: In the adiabatic compression of a fluid within a spherical bubble from radius  $R_i$  to radius  $R_f$ , the temperature rise from  $T_i$  to  $T_f$  is plotted. The temperature rise from room temperature when interactions are included (as in the van der Waals theory) is large compared with the ideal gas case but only if the compression is substantial.

where Eq.(15) has been invoked and the initial volume per particle  $v_i$  determines

$$y = (b/v_i)^{1/3} = (\xi/\Lambda). \quad (23)$$

In Eq.(23),  $b = \xi^3$  and  $v_i = \Lambda^3$ . For argon [37],  $\xi \approx 3.7664 \text{ \AA}$  and for an initial state at room temperature and pressure  $\Lambda \approx 34.356 \text{ \AA}$ . Thus  $y(\text{Ar}) \approx 0.1096$ . A plot of Eq.(22) is shown in FIG.2.

If the final bubble radius  $R_f$  is only (say)  $\approx 10\%$  of the initial radius  $R_i$  [44, 45, 46], then in the van der Waals model the temperature rise can be enormous (say) from room temperature to a final temperature of  $\approx 7.5 \times 10^4 \text{ K}$  at which point the fluid would appear to be incandescent.

#### IV. LAGRANGIAN DYNAMICS

For the purpose of better understanding the role of adiabatic processes in bubble dynamics, we here consider such dynamics from an analytical mechanics viewpoint. The fluid mechanics of a spherical bubble can be described by a Lagrangian of the form

$$L(\dot{R}, R) = \frac{1}{2} \mathcal{M}(R) \dot{R}^2 - \mathcal{U}(R) - \mathcal{U}_{ext}(R), \quad (24)$$

where the external driving potential for the bubble dynamics is denoted by  $\mathcal{U}_{ext}$  and a Rayleigh dissipation function [47]  $\mathcal{F}$  is employed having the form

$$2\mathcal{F}(\dot{R}, R) = \Gamma(R) \dot{R}^2. \quad (25)$$

The dynamical equation of motion reads

$$\frac{d}{dt} \left( \frac{\partial L}{\partial \dot{R}} \right) = \left( \frac{\partial L}{\partial R} \right) - \left( \frac{\partial \mathcal{F}}{\partial \dot{R}} \right). \quad (26)$$

The various terms contributing to Eqs.(24) and (25) will now be explored.

During an expansion or compression of a spherical bubble, the fluid velocity field outside the bubble is

$$\mathbf{v} = \left( \frac{\mathbf{r}}{|\mathbf{r}|^3} \right) R^2 \dot{R} \quad \text{for } (|\mathbf{r}| > R) \quad (27)$$

while the fluid velocity field inside the bubble is

$$\mathbf{v} = \left( \frac{\dot{R}}{R} \right) \mathbf{r} \quad \text{for } (|\mathbf{r}| < R). \quad (28)$$

The (presumed) incompressible flow outside the bubble requires a kinetic energy

$$\begin{aligned} K_{out} &= \frac{\rho}{2} \int_{|\mathbf{r}| > R} |\mathbf{v}|^2 d^3\mathbf{r} \\ &= \frac{\rho}{2} \int_R^\infty \left| \frac{R^2 \dot{R}}{r^2} \right|^2 (4\pi r^2 dr) \\ &= 2\pi \rho R^3 \dot{R}^2, \end{aligned} \quad (29)$$

while the flow of the fluid inside the bubble requires a kinetic energy

$$\begin{aligned} K_{in} &= \frac{M}{2(4\pi R^3/3)} \int_{|\mathbf{r}| < R} |\mathbf{v}|^2 d^3\mathbf{r} \\ &= \frac{3M}{8\pi R^3} \int_0^R \left| \frac{r\dot{R}}{R} \right|^2 (4\pi r^2 dr) \\ &= (3/10)M\dot{R}^2, \end{aligned} \quad (30)$$

where  $M$  denotes the mass of fluid (presumed) conserved within the bubble. From Eqs.(29) and (30) one finds a total fluid flow kinetic energy of

$$K = K_{in} + K_{out} = \frac{1}{2}\mathcal{M}(R)\dot{R}^2 \quad (31)$$

for which

$$\mathcal{M}(R) = \frac{3}{5}M + 4\pi\rho R^3. \quad (32)$$

Eq.(32) establishes the effective mass  $\mathcal{M}(R)$  to be used in the Lagrangian Eq.(24).

For the potential terms in the Lagrangian Eq.(24), one may write a sum of the internal energy of the fluid within the bubble and a term proportional to the surface area

$$\mathcal{U}(R) = N\epsilon \left( s = \frac{S}{N}, v = \frac{4\pi R^3}{3N} \right) + 4\pi\sigma R^2. \quad (33)$$

The external potential may be written in terms of the fluid pressure  $P$  exerted on the bubble by the external fluid; i.e.

$$\mathcal{U}_{ext}(R) = \left( \frac{4\pi R^3 P}{3} \right). \quad (34)$$

Eqs.(32), (33) and (34) completely specify the Lagrangian in Eq.(24).

To compute the dissipation function, we note for the incompressible flow (with viscosity  $\eta$ ) [48] outside the bubble

$$\begin{aligned} 2\mathcal{F}_{out} &= \frac{\eta}{2} \int_{|\mathbf{r}| > R} (\partial_i v_j + \partial_j v_i)^2 d^3\mathbf{r} \\ &= 16\pi\eta R\dot{R}^2, \end{aligned} \quad (35)$$

where Eq.(27) has been invoked. For the flow inside the bubble, only the bulk viscosity  $\zeta$  [48] enters into the computation, i.e.

$$\begin{aligned} 2\mathcal{F}_{in} &= \zeta \int_{|\mathbf{r}| < R} (\text{div}\mathbf{v})^2 d^3\mathbf{r} \\ &= 12\pi\zeta R\dot{R}^2, \end{aligned} \quad (36)$$

where Eq.(28) has been invoked. From Eqs.(35) and (36) one finds a total dissipation function

$$2\mathcal{F} = 2\mathcal{F}_{in} + 2\mathcal{F}_{out} = \Gamma(R)\dot{R}^2 \quad (37)$$

where

$$\Gamma(R) = 16\pi \left( \eta + \frac{3}{4}\zeta \right) R. \quad (38)$$

Eq.(38) establishes the friction coefficient  $\Gamma(R)$  to be used in the dissipation function Eq.(25).

Employing Eqs.(1), (24-26), (32-34) and (38) we find the following equation of motion:

$$\begin{aligned} (1 + \alpha_1)R\ddot{R} + \left( \frac{3\dot{R}^2}{2} \right) + 4(1 + \alpha_2) \left( \frac{\nu\dot{R}}{R} \right) \\ = - \left( \frac{P + (2\sigma/R) - p}{\rho} \right), \end{aligned} \quad (39)$$

where

$$\nu = (\eta/\rho) \quad (40)$$

is the kinematic viscosity of the liquid outside the bubble, and  $\alpha_i$  for  $i = 1, 2$  in Eq.(39) represent the corrections to the usual Rayleigh-Plesset equation [15, 16, 17] due to motions within the bubble. In detail,

$$\alpha_1 = \left( \frac{3M}{20\pi\rho R^3} \right) \quad \text{and} \quad \alpha_2 = \left( \frac{3\zeta}{4\eta} \right). \quad (41)$$

In the usual approximation,  $\alpha_2 \approx 0$ , all of the dissipative heating is from outside the bubble, and the compressions and/or expansions of the bubble are strictly adiabatic. For the internal heating of fluid inside the bubble,  $\alpha_2 > 0$ , the bulk viscosity  $\zeta$  of the fluid inside the bubble will give rise to additional internal heating.

## V. ELECTRONIC EXCITATION OF ATOMS

Let us now further consider the case in which the fluid inside the bubble is made up of argon atoms. We wish to consider the physical chemistry of a simple reaction in which the electron states of argon atoms are excited; i.e.



where  $Ar$  represents an atom in the ground electronic state while  $Ar^*$  represents an atom the atom in an excited electronic state. The concentration of atoms in an excited electronic state is defined as

$$x = \left( \frac{N_{Ar^*}}{N_{Ar} + N_{Ar^*}} \right). \quad (43)$$

The second law of thermodynamics dictates that chemical equilibrium arises when the energy reaches a minimum for constant entropy; i.e.

$$\epsilon(s, v) = \inf_{0 \leq x \leq 1} \tilde{\epsilon}(s, v, x). \quad (44)$$

When the fluid in the bubble is out of chemical equilibrium, Eq.(1) is generalized to

$$d\tilde{\epsilon} = Tds - pdv + \tilde{\mu}dx, \quad (45)$$

where

$$\tilde{\mu} = \mu_{Ar^*} - \mu_{Ar}. \quad (46)$$

If  $x \ll 1$ , then it is sufficiently accurate to use the conventional dilute solution theory wherein

$$\tilde{\mu} = \phi + k_B T \ln x \quad (47)$$

and the activation free energy  $\phi$  is independent of  $x$ . From Eqs.(44) and (45), it follows for chemical equilibrium that  $\tilde{\mu} = 0$ . Eq.(47) then yields the thermal activation formula for concentration

$$x = e^{-\phi/k_B T} \quad (\text{chemical equilibrium}). \quad (48)$$

In thermodynamic mean field theory, the activation free energy may be written in terms of the energy states of a free argon atom as

$$\bar{\phi} = -k_B T \ln \left( \frac{\sum_{E>E_0} w_E e^{-E/k_B T}}{\sum_E w_E e^{-E/k_B T}} \right), \quad (49)$$

where  $w_E$  is the degeneracy of the electronic energy level  $E$  and  $E_0$  is the electronic ground state energy. For argon it is known[49] that the first two electronic excited states are given by

$$\begin{aligned} E(3p^5(^2P_{3/2})4s, J=1) - E_0 &\approx 11.63 \text{ eV} \\ E(3p^5(^2P_{1/2})4s, J=1) - E_0 &\approx 11.82 \text{ eV}. \end{aligned} \quad (50)$$

Including the above electronic states in the computation of the activation free energy in Eq.(49) and employing Eqs.(48) and (22) we plot the concentration of excited states as a function of bubble radius in FIG.3.

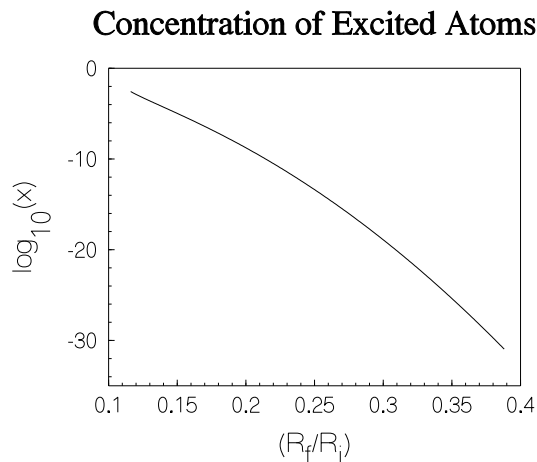


FIG. 3: As bubble containing pure argon is adiabatically compressed from an initial radius  $R_i$  at room temperature and pressure to a final radius  $R_f < R_i$ , the concentration  $x$  of electronically excited argon atoms sharply increases.

## VI. CONCLUSIONS

It has been shown that the adiabatic compression of noble gas atoms gives rise to surprisingly high temperatures. The statistical thermodynamics of the processes may viewed as follows: (i) The entropy of the fluid inside the bubble is proportional to the logarithm of the number of states. (ii) When the bubble is compressed, due to the hard core part of the inter-atomic potential, there is an excluded spatial volume. The compression then eliminates much available spatial volume part of phase space which would in principle lower the entropy. (iii) To keep the entropy constant during the adiabatic compression, the momentum part of phase space must expand as the position part of phase space contracts. Thus there will exist an increased kinetic energy. A high mean kinetic energy (in classical statistical thermodynamics) is equivalent to a high temperature. This explains why an adiabatic bubble compression can lead to such a high temperature.

That noble gas atoms achieve even higher adiabatic compression temperatures than the temperatures which would be achieved with more complex molecular substances has also been explained. The compression lowers the spatial parts of phase space so other parts of phase space must be opened up to keep the entropy constant in an adiabatic process. For a fluid of noble gas atoms, momentum phase space is opened up giving rise to high kinetic energy and high temperature. If the fluid inside the bubble contains molecules with structure, then other parts of phase space can open up to make the lost spatial part of phase space. These include internal tumbling motions of molecules as well as internal vibrational excitations. Not all of the required new regions of phase space involve a rise in center of mass kinetic energy of the molecule which is equivalent to a rise in temperature. Thus, a fluid composed of complex molecules will not exhibit as high a compression induced temperature rise as will a fluid made up of noble gas atoms. From a mathematical point of view, the specific heat  $c_v$  in Eq.(7) describes how much phase space is opened up by a given temperature rise. Each of the internal degrees of freedom in a complex molecule raises  $c_v$ . The higher the specific heat, the lower the temperature rise in Eqs.(10) and (13).

The bubble dynamics has been described through a Lagrangian, including the Rayleigh dissipation function yielding the usual results apart from small correction terms arising from fluid flow within the bubble. Since the energy for fixed entropy enter into the Lagrangian as potential energy terms, the adiabatic compression holds to a high degree of accuracy.

The concentration of electronically excited atoms at compression induced high temperatures can be approximately estimated in mean field theory. One uses an electronic partition function which is strictly valid only for ideal gases. When the fluid is compressed, there will in reality be a shift in electronic energy. The excited electron shells will be somewhat delocalized to neighboring

atoms. In fact, one might well have coherent electron states in such a system giving rise to superradiance during the light emission pulse. However, the situation re-

garding the coherence of sonoluminescent optical pulses is still unclear and requires further elucidation.

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