Supplement to:

A simple consistent explanation of sonoluminescence light emission

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I. CALCULATION OF THE GAS TEMPERATURE

To calculate the time-dependent temperature of the uniform bubble interior, we start with the classical Rayleigh–Plesset equation of bubble dynamics for the radius $R(t)$ of the spherical bubble [1–4]:

$$\rho_l \left(R\ddot{R} + \frac{3}{2}\dot{R}^2 \right) = p_{gas}(R(t)) + p_{vap} - P(t) - P_0 + \frac{R}{c_l}\frac{\mathrm{d}}{\mathrm{d}t}p_{gas}(R(t)) - 4\eta_l\frac{\dot{R}}{R} - \frac{2\sigma}{R}\,, \qquad (1)$$

with the tabulated (e.g. [5]) material constants (liquid viscosity η_l, density ρ_l, speed of sound c_l, and surface tension σ) for the system of a noble gas bubble in water. In (1) p_{gas} is the gas pressure in the bubble, represented here by a van der Waals-type process equation

$$\dot{p}_{gas}(R,t) = \frac{d}{dt}p_{gas}(R(t)) = -\gamma(R,\dot{R},T)\frac{3R^2\dot{R}}{R^3 - h^3}p_{gas}\,, \qquad (2)$$

where h denotes the collective van der Waals hard core radius. An effective polytropic exponent γ is used to describe the thermodynamics of the bubble motion in every time step.

The bubble behaves isothermally when $\gamma \to 1$ and adiabatically when $\gamma \to 5/3$ (for noble gases). The exponent depends on time via the dynamic variables $R, \dot{R}$, and T. Prosperetti [6] has performed a rigorous *analytical* computation of the effective polytropic exponent for the limiting case of linear bubble oscillations. In this case, γ depends only on the Péclet number $Pe = R(t)|\dot{R}(t)|/\chi(t)$, where χ is the thermal diffusivity. Because χ is, in general, a function of density and temperature, it also depends on time. We use Enskog theory [7] to obtain the function $\chi(t) = \chi(R(t), T(t))$ and use Prosperetti's values for $\gamma(Pe)$ to determine the polytropic exponent. While the assumption of linear oscillations is clearly violated for sonoluminescing bubbles, Prosperetti's approach still yields physically reasonable values, namely, $\gamma \to 1$ (isothermality) for low Pe and $\gamma \to 5/3$ (adiabaticity) for $Pe \to \infty$. The latter case occurs only in the immediate vicinity of the bubble collapse. During the rest of the oscillation cycle (i.e., almost all the time) the bubble behaves isothermally.

Now we use the van der Waals equation of state to translate (2) into an ODE for $T(t)$,

$$\dot{T} = -\left[\gamma(Pe) - 1\right] \frac{3R^2\dot{R}}{R^3 - h^3} T \,. \tag{3}$$

Solving the coupled system of ODEs (1) and (3) yields the time series of temperature used to evaluate the photon absorption coefficients and the resulting light emission. The maximum temperatures achieved usually lie between 20 000 K and 30 000 K and show only moderate variations over the parameter range of sonoluminescing bubbles.

II. PHOTON ABSORPTION COEFFICIENTS IN A WEAKLY IONIZED GAS

All absorption processes described here occur only when at least a small number of ions and/or free electrons are present (all absorption coefficients are linearly or quadratically proportional to the degree of ionization α). The Saha equation gives α explicitly as

$$\alpha[T] = \left(\frac{2\pi m_e k_B T}{h^2}\right)^{3/4} \left(\frac{2u_+}{nu_0}\right)^{1/2} \exp\left(-\frac{E_{ion}}{2k_B T}\right) , \tag{4}$$

with the electron mass m_e, Boltzmann and Planck constants k_B and h, and statistical weights u_+, u_0 for the ground states of the ion and the neutral gas atom, respectively. We follow the hydrogen-like atom model of [8] and set $2u_+/u_0 = 1$. Considering the large ionization energies $E_{ion} = 15.8\,eV$ for Ar or $12.1\,eV$ for Xe, it is apparent from the exponential factor $\exp(-E_{ion}/2k_BT)$ in (4) that at maximum temperatures of $\approx 2-3\,eV$ $(20\,000 - 30\,000\,\mathrm{K})$ ionization is rare.

When evaluating different contributions to photon absorption from the literature [8], we find three terms to be of vital importance in at least part of the parameter range of sonoluminescence explored today in experiment. They are described in the following subsections.

A. Free-free transitions of electrons near ions

At high or moderate degrees of ionization, photons will predominantly be absorbed by free electrons, where an ion acts as a third partner to satisfy conservation of both energy and momentum. The absorption coefficient for this process is [8]

$$\kappa_\lambda^{(i)}[T] = \frac{4}{3}\left(\frac{2\pi}{3m_e k_B T}\right)^{1/2} \frac{Z^2 e^6 \lambda^3}{(4\pi\varepsilon_0)^3 h c^4 m_e} \left(\alpha[T]\right)^2 n^2\,, \tag{5}$$

with an effective charge Z of ions (here, $Z = 1$), the elementary charge e, the speed of light c, and the vacuum permeability ϵ_0. The number density of electrons (and therefore of ions) is denoted by n. The inverse (emission) process corresponding to this absorption is the well-known bremsstrahlung of electrons in the field of ions.

B. Free-free transitions of electrons near neutral atoms

As α is small for typical SBSL situations, there are far more neutral atoms than ions present in the bubble. Therefore, collisions of electrons with neutral atoms are important despite their much smaller interaction cross section (compared to electron-ion collisions).

This *transport scattering cross section* [9] depends on the energy of the electron; for argon we have with good accuracy the linear relation [10]

$$\sigma_{tr}(E_e) \approx c_{tr} E_e + d_{tr} \,, \tag{6}$$

for the relevant thermal electron energies E_e (a few eV) here, with constants $c_{tr} \approx 1.6 \cdot 10^{-20} m^2/eV$ and $d_{tr} \approx -0.6 \cdot 10^{-20} m^2$. Using this expression and averaging the photon absorption coefficient for this process over the Maxwell-distributed electron energies [8], we obtain

$$\kappa_\lambda^{(ii)}[T] = \frac{e^2}{\pi\varepsilon_0} \frac{(2k_B T)^{9/4} n^{3/2}}{h^{3/2} c^3 m_e^{3/4} \pi^{3/4}} \lambda^2 \left(c_{tr} + \frac{d_{tr}}{3k_B T} \right) \exp\left(-\frac{E_{ion}}{2k_B T} \right) . \tag{7}$$

Here, the inverse process is *neutral bremsstrahlung*, i.e., photon emission from electrons colliding with neutral atoms. When computed numerically, the size of this contribution is indeed comparable to $\kappa_\lambda^{(i)}$ in a wide range of parameters in the SBSL range.

C. Bound-free transitions of electrons

Finally, we consider ionization of electrons through absorption of photons. In the optical wavelength regime we are interested in, an ionization from the ground state $j = 1$ of a noble gas atom is impossible. However, the gap between the ground state and the first excited state $j = 2$ is so large that at least for a range of λ all levels $j \geq 2$ can participate in the process. More precisely, for every λ there exists a $j = j^*$ which is the smallest j for which the j-th energy level $E_j = E_{ion}/j^2$ of a hydrogen-like atom is smaller than hc/λ. Electrons on all levels with $j \geq j^*$ can then be ionized by a photon of wavelength λ and their contribution must be summed up to yield the absorption coefficient

$$\kappa_\lambda^{(iii)}[T] = \frac{64\pi^4}{3\sqrt{3}} \frac{e^{10} n}{(4\pi\varepsilon_0)^5 h^6 c^4} \lambda^3 \sum_{j=j^*}^{\infty} \frac{1}{j^3} \exp\left(-\frac{E_{ion} - E_j}{k_B T} \right) . \tag{8}$$

Above the first excited state, the level spacing becomes so narrow [11] that a continuum model is appropriate [8]. Thus, (8) can be simplified and added to $\kappa_\lambda^{(i)}$ to obtain a corporate absorption coefficient for both processes involving ions,

$$\kappa_\lambda^{(i)+(iii)}[T] = \frac{16\pi^2}{3\sqrt{3}} \frac{e^6 k_B T n}{(4\pi\varepsilon_0)^3 h^4 c^4} \lambda^3 \exp\left(-\frac{E_{ion} - hc/\max\{\lambda, \lambda_2\}}{k_B T} \right) . \tag{9}$$

As λ decreases, more and more electron energy levels become available for ionization, so that κ_λ^{ion} increases. But for photons with energies higher than that of the first excited level $E_2 \equiv hc/\lambda_2$, there are no additional energy levels available (the ground state lying too low to be reached). Therefore, the ionic absorption at wavelength λ_2 will not increase for $\lambda < \lambda_2$, but instead decrease with decreasing λ, as does the neutral absorption (7). The resulting maximum in absorptivity is reflected by an emissivity maximum in the observed spectrum, whose location is very close to λ_2 (see Fig. 1 of the main text).

Adding the contributions $\kappa_\lambda^{(i)+(iii)}$ and $\kappa_\lambda^{(ii)}$, we arrive at the quantitative formula for the total absorption coefficient $\kappa_\lambda[T(t)]$ we sought for.

III. LIGHT EMISSION INTENSITY

Emitted radiation that has traveled a distance s through a bubble (radius R) of spatially uniform temperature T (and therefore uniform $\kappa_\lambda[T(t)]$) shows the spectral intensity [8, 12]

$$I_\lambda(s,t) = I_\lambda^{Pl}[T(t)] \left(1 - \exp\left(-\kappa_\lambda[T(t)]s\right)\right) , \quad 0 < s < 2R . \tag{10}$$

This quantity represents energy per unit time, wavelength interval, solid angle, and projected surface area. It becomes the Planck intensity $I_\lambda^{Pl}(t)$ for $\kappa_\lambda R \to \infty$, but in the case of sonoluminescing bubbles this product is small compared to one. Integrating over the projected surface of the bubble and all solid angles, we arrive at the total emitted power from the bubble per wavelength interval at wavelength λ,

$$P_\lambda(t)d\lambda = 4\pi^2 R^2 I_\lambda^{Pl}[T(t)] \left(1 + \frac{\exp\left(-2\kappa_\lambda R\right)}{\kappa_\lambda R} + \frac{\exp\left(-2\kappa_\lambda R\right) - 1}{2\kappa_\lambda^2 R^2}\right) d\lambda\,. \tag{11}$$

For $\kappa_\lambda R \gg 1$, this becomes the power emitted from an ideal black body (Planck emitter), whereas for $\kappa_\lambda R \ll 1$ we find a volume emitter radiation of the form

$$P_\lambda(t)d\lambda = 4\pi\kappa_\lambda I_\lambda^{Pl}[T(t)]\frac{4\pi R^3}{3}d\lambda = \frac{4}{3}\kappa_\lambda R\, P_\lambda^{Pl}(t)d\lambda\,. \tag{12}$$

The Planck emission strength is thus reduced in proportion to the value of $\kappa_\lambda R$. As this limit is usually valid in typical SBSL parameter regimes, it explains the smaller photon yields as compared to an ideal black body calculation. The sensitive dependence of all contributions to κ_λ on T via the degree of ionization (4) accounts for the shortness and wavelength independence of sonoluminescence pulses, as pointed out in the main text of the paper.

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