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Single-bubble sonoluminescence from noble gases

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Single-bubble sonoluminescence (SBSL) from noble gases in water is studied theoretically in order to clarify the reason of the distinguished feature that the luminescence is strong for all noble gases, while the other systems of cavitation luminescence are greatly enhanced by the presence of the heavy noble gas(xenon). It is clarified that in spite of the larger thermal conductivity of lighter noble gases the maximum temperature in a SBSL bubble of lighter noble gases is higher due both to the segregation of water vapor and noble gas inside a SBSL bubble and the stronger acoustic drive of a SBSL bubble of lighter noble gases.

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Single-bubble sonoluminescence (SBSL) is a light emission phenomenon from a stably oscillating bubble in liquid irradiated by a standing ultrasonic wave [1]. The light is emitted at each bubble collapse as a pulse. In 1991, Barber and Putterman [2] reported that the pulse width of SBSL is extremely small (~50 ps). Since the report, many researchers have studied the phenomenon both theoretically and experimentally [3].

In 1999, Weninger, Camara, and Putterman [4] reported that there are two categories in the luminescence from bubbles: one is the systems which are greatly enhanced by the presence of xenon, and the other is single-bubble sonoluminescence in water which is strong for all noble gases. The first category includes multibubble sonoluminescence [5–7], single-bubble sonoluminescence in nonaqueous liquids [8], sonoluminescence from a hemispherical bubble on a solid surface [9], and the luminescence associated with flow-induced cavitation [10].

Multibubble sonoluminescence (MBSL) in the first category is the luminescence from hundreds or thousands of bubbles in liquid irradiated by a strong ultrasound [5–7]. It is widely accepted [7,11,12] that the maximum temperature inside a MBSL bubble is higher for heavier noble gases due to the lower thermal conductivity: a bubble is less cooled by the thermal conduction from the heated interior of the bubble to the surrounding liquid when the thermal conductivity is lower. Thus, in MBSL, xenon bubbles are the brightest and helium bubbles are the dimmest [5–7].

In the present study, computer simulations of collapses of noble-gas bubbles are performed under conditions of SBSL in water in order to study the reason of the distinguished feature of SBSL in water that it is strong for all noble gases.

In 1999, Storey and Szeri [13] predicted theoretically that a gas mixture is mildly segregated inside a collapsing bubble under a condition of SBSL in water by the thermal and pressure diffusion. The lighter gas molecules are driven to the bubble center and the heavier gas molecules are driven to the bubble wall. Thus inside a xenon bubble, water vapor is driven to the bubble center and xenon is driven to the bubble wall. On the other hand, inside a helium bubble, water vapor is driven to the bubble wall and helium is driven to the bubble center. Inside an argon bubble, no significant mixture segregation takes place because molecular mass of argon is not so different from that of water vapor [14].

Although the mixture segregation inside a SBSL bubble is mild, it affects the bubble dynamics through the change of the rate of vapor condensation at the bubble wall. At the bubble collapse, vapor condenses at the bubble wall due to the increase of pressure inside a bubble [15]. The rate of vapor condensation at the bubble wall is nearly proportional to the vapor pressure at the bubble wall [15]. By the mild mixture segregation, the vapor pressure at the bubble wall changes and accordingly the rate of vapor condensation changes.

In the present computer simulations, the mild mixture segregation is taken into account by simple model equations. For a xenon bubble, $p_{\mathrm{Xe},B}=\overline{p}_{\mathrm{Xe}}+(p_g-\overline{p}_{\mathrm{Xe}})n_{\mathrm{Xe}}/n_t$, $p_{v,B}=p_g-p_{\mathrm{Xe},B}$, where $p_{\mathrm{Xe},B}$ is the partial pressure of xenon at the bubble wall, $\overline{p}_{\mathrm{Xe}}$ is the spatially averaged partial pressure of xenon, p_g is the pressure inside a bubble, n_{Xe} and n_t are the number of xenon atoms and the total number of particles inside a bubble, respectively, and $p_{v,B}$ is the vapor pressure at the bubble wall. The equations are derived by the following requirements: when the bubble content is mostly xenon, $p_{\mathrm{Xe},B}\sim p_g$, and when the content is mostly vapor, $p_{\mathrm{Xe},B}\sim\overline{p}_{\mathrm{Xe}}$. The obvious requirement $\overline{p}_{\mathrm{Xe}}<p_{\mathrm{Xe},B}<p_g$ is automatically satisfied by $p_{\mathrm{Xe},B}=\overline{p}_{\mathrm{Xe}}+\alpha(p_g-\overline{p}_{\mathrm{Xe}})$, where $0<\alpha<1$. The above requirements are satisfied by $\alpha=n_{\mathrm{Xe}}/n_t$. Adding the condition that $p_{\mathrm{Xe},B}+p_{v,B}=p_g$, the model equations are derived. In the computer simulations, the model equations are used from 0.01 μs before the time of the minimum bubble radius (R_{min}) according to the results in Ref. [13]. The model equations yield values of $p_{\mathrm{Xe},B}/\overline{p}_{\mathrm{Xe}}$

TABLE I. The effective black-body temperature (T_{eff}) and the energy of the emitted light per SBSL pulse estimated from the spectra reported by Hiller and co-workers [3,29]. The degree of saturation of the gas in water is 0.002 for He and Ar and 0.004 for Xe. The values in brackets are for the degree of saturation of 0.2. The frequency of ultrasound is 33 kHz and the ambient liquid temperature is 24 °C.

Gas	He	Ar	Xe
T_{eff}	15 000 K	15 000 K	10 000 K
	(20 000 K)	(15 000 K)	(10 000 K)
Energy per SBSL pulse	0.4 pJ	0.4 pJ	1.2 pJ
	(0.1 pJ)	(0.1 pJ)	(0.4 pJ)

TABLE II. The calculated results of a SBSL bubble for various amplitudes of ultrasound p_a when Ar is dissolved in 24 °C water with the degree of saturation of 0.002. The frequency of ultrasound is 33 kHz. R_0 is the ambient bubble radius that is calculated by the Eller-Flynn formula [37]. T_{max} is the maximum bubble temperature, ''energy'' is that of the emitted light per pulse, ''pulse width'' is that of the emitted light, ''mechanism'' is that of the light emission, ''atom bremss.'' is electron-atom bremsstrahlung, and ''rad. rec.'' is radiative recombination.

p_a	1.44 bar	1.47 bar	1.52 bar
R_0	4 μm	4.5 μm	5 μm
T_{max}	14 000 K	14 000 K	15 000 K
Energy	0.3 pJ	0.5 pJ	0.9 pJ
Pulse width	110 ps	120 ps	130 ps
Mechanism	atom bremss. rad. rec.	atom bremss. rad. rec.	atom bremss. rad. rec.

$\sim$1.1 at 0.01 μs before R_{min}, which is in agreement with the results in Ref. [13].

Similarly for a helium bubble, the following model equations are used: $p_{v,B}=\overline{p_v}+(p_g-\overline{p_v})n_{H_2O}/n_t$, $p_{He,B}=p_g-p_{v,B}$, where $\overline{p_v}$ is the spatially averaged partial pressure of water vapor, n_{H_2O} is the number of water vapor molecules inside a bubble, and $p_{He,B}$ is the partial pressure of helium at the bubble wall.

For a bubble collapse under a condition of SBSL, there are two theoretical models: one is the shock-wave model [16–19] that a spherical shock-wave develops inside a collapsing bubble, and the other is the quasiadiabatic compression model [15,20] that no shock-wave is formed inside a bubble and a bubble is almost uniformly heated by a quasiadiabatic compression, where ''quasi'' means that appreciable thermal conduction takes place between a bubble and the surrounding liquid. In 1998, Cheng and co-workers [21,22] clarified by computer simulations of the fundamental equations of fluid dynamics inside a collapsing bubble that no shock-wave develops inside a SBSL bubble and the spatial variation of pressure and temperature inside a bubble is only a few tens percent. The reason of no shock formation was theoretically clarified by Vuong, Szeri, and Young in 1999 [23]. Thus, in the present computer simulations of bubble collapses, a quasiadiabatic compression model is used [15].

In the model [15], the effect of thermal conduction both inside and outside a bubble, that of nonequilibrium evaporation and condensation of water vapor at the bubble wall, and that of chemical reactions inside a heated bubble including the thermal dissociation of water vapor are taken into account. In the present study, radiative processes inside a bubble are also simulated by the model described in Ref. [24]. The radiative processes considered are bremsstrahlung of electrons, radiative recombination of electrons and ions, and radiative attachment of electrons to neutral atoms. For bremsstrahlung, there are two mechanisms, electron-atom bremsstrahlung [25] and electron-ion bremsstrahlung [26], that are the light emissions from free electrons accelerated in a field of a neutral atom and that of a positive ion, respectively. In the present simulations, the number of free electrons inside a bubble is calculated by the Saha equation [27], taking into account the reduction of ionization potentials of gases by the extreme high density inside a SBSL bubble [28].

The present simulations are performed under the condition of the experiment reported by Hiller and co-workers [3,29]. In 1994, Hiller *et al.* [29] reported the SBSL spectra from various noble gases. Each spectral shape reported [3,29,30] can be fitted by the black-body formula (Planck function). In other words, an effective black-body temperature exists for each spectrum. In Table I, the effective black-body temperature (T_{eff}) and the energy of the emitted light per pulse estimated from the reported spectra [3,29] are listed. It should be noted that in the experiment [3,29] nitrogen is also dissolved in water in addition to noble gases. However, according to Lohse *et al.* [31], a SBSL bubble consists of noble gases because nitrogen molecules inside a bubble change to the soluble species such as NO_x and HNO_x by chemical reactions and dissolve into the surrounding water. The hypothesis of Lohse *et al.* [31] has been confirmed experimentally [32,33].

According to the recent theories [24,34,35], SBSL is not the black-body radiation. Nevertheless, the theories [24,35,36] predict that the spectral maximum of SBSL shifts towards shorter wavelengths as the bubble temperature increases, which is the same trend as that of the black-body radiation. Thus, it is concluded from Table I that the maximum bubble temperature is higher for lighter noble gases, in contrast to the MBSL case. In the present Rapid Communication, the difference between SBSL and MBSL is addressed.

In Table II, the calculated results of an Ar bubble are

TABLE III. The calculated results of a SBSL bubble for various amplitudes of ultrasound p_a when Xe is dissolved in 24 °C water with the degree of saturation of 0.004. The frequency of ultrasound is 33 kHz. R_0 is calculated by the Eller-Flynn formula [37]. For T_{max}, the values when the mild mixture segregation is neglected are also listed.

p_a	1.40 bar	1.43 bar	1.47 bar
R_0	5 μm	5.5 μm	6 μm
T_{max}	9 000 K	10 000 K	10 000 K
(without mix.seg.)	(12 000 K)	(13 000 K)	(13 000 K)
Energy	0.2 pJ	0.5 pJ	1.0 pJ
Pulse width	170 ps	180 ps	190 ps
Mechanism	atom bremss.	atom bremss.	atom bremss.

TABLE IV. The calculated results of a SBSL bubble when He is dissolved in 24 °C water. The frequency of ultrasound is 33 kHz. For T_{max}, the value when the mild mixture segregation is neglected is also listed.

p_a	1.7 bar
R_0	3 μm
T_{max}	16 000 K
(without mix. seg.)	(9 000 K)
Energy	0.1 pJ
Pulse width	80 ps
Mechanism	atom bremss.

listed for various amplitudes of ultrasound when the frequency of ultrasound is 33 kHz and the ambient liquid temperature is 24 °C. The ambient bubble radius (R_0) is calculated by the condition of the balance of gas diffusion between the bubble and the surrounding liquid (the Eller-Flynn formula) [37]: $\int_0^{T_b} R^4((p_g - p_v) - p_\infty c_i/c_0)dt = 0$, where T_b is the period of ultrasound, R is the bubble radius, $(p_g - p_v)$ is the partial pressure of noncondensable gas [the total pressure (p_g) minus the vapor pressure (p_v)] inside a bubble, p_∞ is the ambient pressure (1 atm), c_i is the actual gas concentration in water apart from the bubble, and c_0 is the solubility of the gas in water. From Table II, it is seen that the maximum bubble temperature is nearly independent of the driving pressure.

In Table III, the calculated results of a Xe bubble are listed. For the maximum bubble temperature, the calculated values when the mixture segregation is neglected are also listed. As in the case of Ar, the ambient bubble radius (R_0) is calculated by the Eller-Flynn formula [37]. It is seen that the maximum bubble temperature is nearly independent of the acoustic amplitude and lower than that of an Ar bubble. It should be noted that even under the same acoustic amplitude (p_a) and ambient radius (R_0), the maximum temperature of a Xe bubble is lower than that of an Ar bubble due to the effect of the mixture segregation. For example, when $p_a = 1.47$ bar and $R_0 = 4.5\ \mu$m, the maximum bubble temperature of Xe is 10 000 K with the mixture segregation (15 000 K without the mixture segregation), while that of Ar is 14 000 K.

Now we discuss the reason of the reduction of the maximum bubble temperature by the mild mixture segregation inside a Xe bubble. By the mild accumulation of xenon near the bubble wall, the vapor pressure at the bubble wall decreases, which results in the decrease of the rate of vapor condensation at the bubble wall at the bubble collapse. As a result, the amount of water vapor trapped inside a bubble and dissociated by the high temperature increases. It decreases the maximum bubble temperature considerably due to the increase of the endothermal heat of dissociation. It should be noted here that the vapor condensation at the bubble wall is a nonequilibrium process since the bubble collapse is so fast [38].

For a He bubble, the maximum temperature is lower than that of an Ar bubble (~15 000 K) for any driving pressure if the ambient bubble radius is calculated by the Eller-Flynn formula. Thus, it is expected that a He bubble does not obey the Eller-Flynn formula. In Table IV, the calculated result of a He bubble when $p_a = 1.7$ bar and $R_0 = 3\ \mu$m is shown. Under the condition, the maximum temperature is higher than that of an Ar bubble. On the other hand, if the mild mixture segregation is neglected, the calculated temperature is always much lower than that of an Ar bubble. It suggests that the mild mixture segregation indeed takes place inside a He bubble.

The increase of the maximum temperature in a He bubble by the mild mixture segregation is caused by the increase of the amount of vapor condensation at the bubble wall, which results in the decrease of the amount of vapor trapped inside a bubble and dissociated by the high temperature. The maximum bubble temperature increases due to the decrease of the endothermal heat of dissociation. The higher maximum bubble temperature than that of Ar is not only due to the mild mixture segregation but also to the stronger acoustic drive. At the same acoustic amplitude, the bubble temperature of He is lower than that of Ar. For example, when $p_a = 1.47$ bar and $R_0 = 4.5\ \mu$m, T_{max} of He is 9 000 K even with the mixture segregation, while that of Ar is 14 000 K.

In conclusion, inside a SBSL bubble in water, water vapor and noble gas (helium or xenon) are mildly segregated. As a result, the maximum temperature in a SBSL bubble of lighter noble gases becomes higher with the fact that it is driven by stronger ultrasound. It results in the distinguished feature of SBSL in water that it is bright for all noble gases, with the fact that the ionization potential of heavier noble gases is lower.

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