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Electron-atom bremsstrahlung and the sonoluminescence of rare gas bubbles

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The bremsstrahlung spectra of electrons interacting with neutral rare gas atoms are calculated for temperatures from 5000 to 40 000 K. The calculations were previously shown to be in agreement with measurements of such spectra recorded under a variety of conditions. The computational results are compared with measured sonoluminescence (SL) spectra of rare gas bubbles. For the heavier rare gases, computed intensities and spectral line shapes compare favorably with the measurement at electron ''temperatures'' of roughly 20 000 K. The agreement suggests that electron–neutral-atom bremsstrahlung may be a principal mechanism of the light emission of rare gas SL bubbles if a weakly ionized SL environment may be assumed.
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I. INTRODUCTION

It has long been known that, in the tensile phase of (nonlinear) soundwaves in water, cavitation occurs [1,2]. During the subsequent compression phase, the cavities (''bubbles'') collapse, so that their gas content is compressed. As a consequence, the temperature of the gas is increased, either adiabatically or by a shockwave mechanism. It has been observed that the cavities collapse at speeds that are roughly four times the speed of sound, so that a spherical, convergent shockwave is driven [3,4]. The shock front is reflected at the bubble center, momentarily creating ($\approx 10^{-10}$ s) a region of high gas density ($\approx 10^{22}$ particles per cm^3) and high temperature which may emit a flash of light (''sonoluminescence,'' SL). Estimates of the peak ''temperatures'' vary widely; they range from several thousand to millions of K. Temperature estimates at the low end of this range have been convincingly demonstrated for SL in organic liquids [5,6], while the higher temperatures were claimed for SL in water [4]. Therefore, water has been considered by some SL investigators to be a very special liquid. However, it remained unclear what specific properties of water would make that liquid so special.

The nature of the light emitting processes of SL is not understood. Spectra of SL bubbles in water, in which various gases were dissolved in controlled ways, have been recorded [7–11]. According to such studies, rare gas bubbles are much brighter than bubbles filled with other gases, especially if the water temperatures are close to freezing (presumably so that the H_2O vapor pressure is minimized) [4]. SL with pure diatomic gases, and with pure molecular gases in general, glows faintly or not at all. The fact that the SL of water saturated with air was already observed more than 60 years ago [12] is now thought to be due to the $\approx 1\%$ argon content of air: strongly forced air bubbles are believed to break down the common molecular gases, so that after a short time more or less pure argon exists in the bubbles [13]; nitrogen bubbles without a trace of argon do not glow. In any case, it is clear that the SL emission intensities vary with the nature (and the concentration) of the gas dissolved in the water, and also with the water temperature.

The SL spectra in water are continuous, without any discernible line or band structures in the spectral ''window'' of water (i.e., at wavelengths between 200 and 700 nm), regardless of the nature of the gases employed [7–11]. Actually, apart from the striking intensity variations between the various gases, the spectral profiles seem to be somewhat independent of the nature of the gas: a near exponential falloff with increasing wavelengths ($\lambda \to 700$ nm), and a broad maximum at short wavelengths. In some cases such a maximum seems to fall just outside of that window ($\lambda < 200$ nm), where, however, it cannot be observed.

In a recent paper with the title ''Defining the unknowns of SL'' [4], the various known attempts to understand the nature of the light emitting processes of SL are briefly discussed and then discarded—there are problems with all of them. For the purpose of discussion, however, electron-ion bremsstrahlung of a hypothetical, optically dense plasma was mentioned as the most likely successful candidate, in spite of certain inconsistencies of that model with the known facts. Specifically, the emission from such a plasma should probably depend not so much on whether the plasma was generated from, say argon or nitrogen gas, but the measurements certainly show dramatic intensity differences. Moreover, no trace of an afterglow (or recombining plasma) has yet been discovered, and the existence of a dense plasma has certainly not been demonstrated; such a plasma may actually not exist under SL conditions. In other words, the SL emission processes are still unclear, and so are certain SL temperature estimates which were based on Planck's blackbody formula: below we will show some evidence which suggests that the SL source may not be optically dense, so that Planck's formula may not be applicable.

For desirable progress in the understanding of SL emission, we think it worthwhile to consider what contributions the familiar emission processes known from the studies of neutral and weakly ionized, hot environments could make, rather than focusing on the emission of dense plasmas. Specifically, here we are interested in the spectra arising from electron–neutral-atom collisions. Such emission is also called bremsstrahlung, as is the emission from electron-ion collisions, but the former is important at lower temperatures;

both bremsstrahlung processes are similarly efficient in generating light. Other processes, e.g., supramolecular emission, may also be important under SL conditions [14], especially in molecular gases; these will be considered in detail elsewhere.

The absorption and emission spectra of electron–rare-gas atom collisions are well known from theory [15–19]. A great diversity of experimental studies of such spectra also exists. We mention specifically drift tube studies of the light emitted by electrons drifting in argon gas in response to an external electric field [20]. Moreover, spectroscopic measurements of shock-heated rare gases were reported, with temperatures from 8000 to 15 000 K [21]; studies of partially ionized, ''hot'' gases exist [22]; the emission of positive columns of rare-gas discharges has been studied [23–25]; and laser-induced gas breakdown studies have also provided relevant information [26]. Many of these spectroscopic studies include careful comparisons with the theory, which is well established. In most cases close agreement of theory and measurement was observed [15,20]. The works quoted above are concerned mainly with electron–rare-gas atom spectra; a comprehensive bibliography of related work is given elsewhere [27]. We think that reliable predictions of such spectra to be expected under SL conditions can be made, if a weakly ionized environment may be assumed. The results should be of interest for estimates of the significance of the neutral bremsstrahlung contributions to SL, for new temperature estimates, and for a possible reduction of the number of the many unknowns that at present still plague SL research.

II. THEORY

The spectral intensity $I(\omega)d\omega$ in units of energy per time and per volume is given by [15,16,20]

$$I(\omega)d\omega = N_e N\hbar\left(\int_{\hbar\omega}^{\infty} v\,Q_{\mathrm{ff}}(\epsilon,\omega)f(\epsilon)d\epsilon\right)d\omega. \tag{1}$$

Here N_e and N are the number densities of the electrons and neutral atoms, respectively; ϵ and v stand for the kinetic energy and velocity of the electron; $f(\epsilon)$ is the normalized distribution function of the electron energies (which we will assume to be Maxwellian, with temperature T),

$$\int_0^{\infty} f(\epsilon)d\epsilon = 1;$$

and $Q_{\mathrm{ff}}(\epsilon,\omega)$ is the cross section for emitting a photon of angular frequency $\omega = 2\pi c\nu$ in a free-free transition [28],

$$Q_{\mathrm{ff}}(\epsilon,\omega) = \frac{8a_0}{3\pi\hbar c}\left(\epsilon - \frac{\hbar\omega}{2}\right)\left(1 - \frac{\hbar\omega}{\epsilon}\right)^{1/2} Q_m(\epsilon). \tag{2}$$

This formula was shown to reproduce measured e-Ar bremsstrahlung spectra reliably, within ±30%, for mean electron energies from 1.2 to 5.4 eV [20]. Several of the diverse measurements quoted above were also shown to be consistent with the theory employed here, or with an equivalent theory, but not always with comparable precision. In Eq. (2), a_0 is Bohr's radius of the hydrogen atom; c is the speed of light; $\hbar$ is Planck's constant, divided by 2π; and $Q_m(\epsilon)$ is the cross section for momentum transfer, which is well known [20,29,30]. Strictly speaking, Eq. (2) was obtained with the assumptions of small photon energies, $\hbar\omega \ll \epsilon$. However, Eq. (2) has actually been shown to be good up to photon energies near threshold, $\hbar\omega \approx \epsilon$ [20].

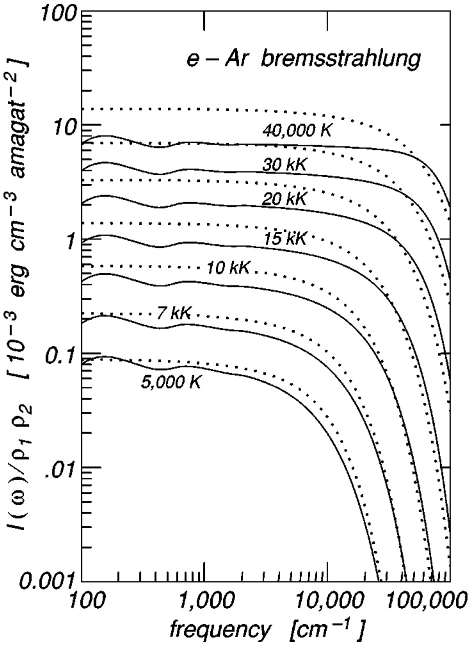

FIG. 1. The emission spectra, normalized by the densities of the electrons and neutral atoms, of collisional e-Ar pairs at seven temperatures, from 5000 to 40 000 K; quantum line shape computations: solid curve; calculations based on Eqs. (1) and (2): dashed line.

It is well known that, alternatively, one may compute the bremsstrahlung spectra from first principles. We have repeated such calculations, using the same modified Hartree-Fock-Slater potential and general procedures that were used previously for the purpose [15]. The radial Schrödinger equation was integrated using the Numerov algorithm with radial step sizes near zero radius of 10^{-4} atomic length units.

III. RESULTS

Figure 1 shows the calculated bremsstrahlung spectra arising from collisions of electrons with argon atoms. The solid curves present the results of the quantum line-shape calculations; the dashed curves are based on empirical collisional cross sections [Eq. (1)]. Reasonable consistency is observed, especially at the lower temperatures. A ''white'' continuum is observed, with intensities nearly independent of frequency, up to a point where the products of optical frequency and the duration of a collision are of the order of unity, in the visible or near ultraviolet region of the spectrum. At such high frequencies, the bremsstrahlung spectrum falls off rapidly. Intensities increase sharply with increasing temperature.

These e-Ar spectra are quite characteristic of all electron–neutral-atom spectra of the rare gases. The most striking differences among the rare gases are the higher intensities of e-Kr and e-Xe collisions, and the lower intensities of e-He and e-Ne interactions (relative to Fig. 1), as we will see below.

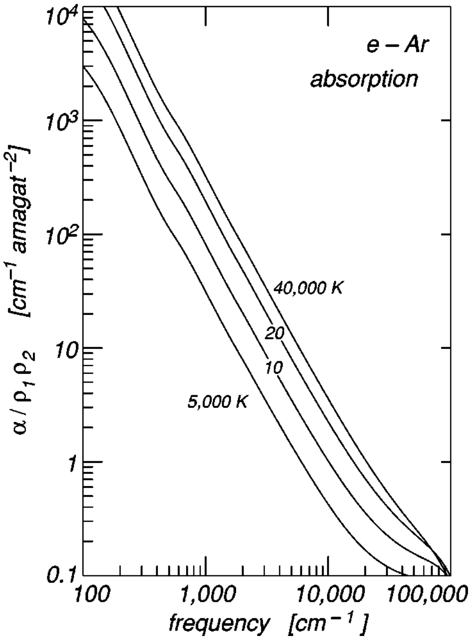

FIG. 2. The absorption spectra, normalized by the densities of electrons and neutral atoms, of collisional e-Ar pairs at four temperatures from 5000 to 40 000 K; from quantum line shape calculations.

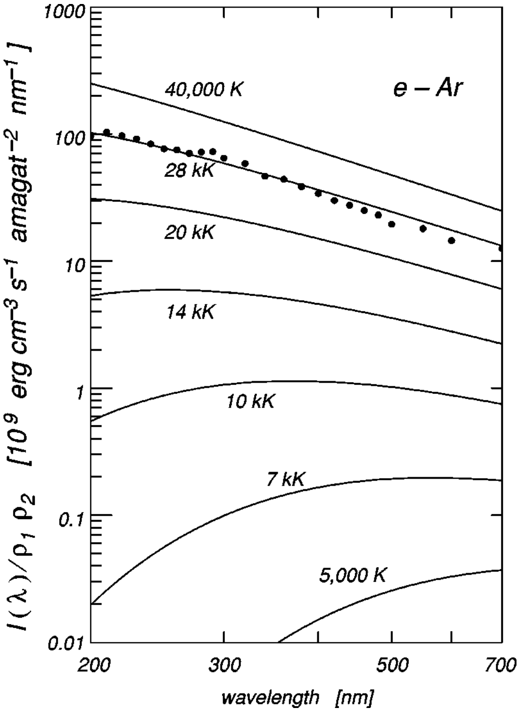

FIG. 3. The e-Ar emission spectra, from Fig. 1, in the spectral window of liquid water, as a function of wavelength, for seven temperatures from 5000 to 40 000 K. A measured SL spectral profile of argon bubbles is also shown (dots; from Ref. [4]).

We note that, in the figures, densities $\varrho = N/N_L$ are expressed in units of Loschmidt's number $N_L = 2.68675\times10^{19}$ particles per cm^{-3}, the density of an ideal gas at standard temperature (0 °C), and pressure. For our purpose, that choice is practically equivalent to the amagat unit widely used when dealing with dense gases.

Figure 2 shows the associated absorption coefficient α as function of frequency, normalized by the densities of the electrons and atoms. The results are based on quantum line shape calculations. The absorption coefficient α is related to the data shown in Fig. 1, according to [15]

$$\alpha(\omega) = \frac{\pi^2 c^2}{\hbar\omega^3}[\exp(\hbar\omega/kT) - 1]I(\omega). \tag{3}$$

Here k is Boltzmann's constant. Knowledge of the absorption coefficient is important for estimating the optical density of the bremsstrahlung light source; the mean free path of a photon is given by the reciprocal absorption coefficient, $\lambda_{\text{ph}} = 1/\alpha$. In the log-log scale, we note a nearly linear falloff with increasing frequency over several orders of magnitude. Again, the absorption spectrum shown here for e-Ar pairs is characteristic of all the rare gases, with intensity variations among the different rare gases commensurate with the variations of the emission.

In order to compare the calculated spectral profiles (Fig. 1) with an appropriate measured SL line shape, in Fig. 3 we plot that segment of the data with wavelengths λ from 200 to 700 nm, the spectral window of liquid water. The intensity $I(\lambda)$ is related to $I(\omega)$ [Eq. (1)] according to $I(\omega)d\omega = I(\lambda)d\lambda$. Also shown is a recorded line shape (dots) [8] of SL spectrum of argon bubbles, which appears to be of a similar shape to the computed profiles at a temperature of roughly 20 000 K. For this comparison of theoretical and measured spectral shapes, we have arbitrarily shifted the measured profile vertically, for easy comparison with the calculated profiles for temperatures around 20 000–30 000 K. (At this point a comparison of calculated and measured intensities is not intended, but will be made below.) Computed and measured spectral profiles are similar.

Similarly, Figs. 4–7 compare measured SL profiles of krypton, xenon, helium, and neon [4] with our calculations. For e-Kr and e-Xe pairs, good agreement is observed at temperatures around 14 000 K. For e-He and e-Ne pairs, on the other hand, the logarithmic slopes of the measured profiles seem to be somewhat steeper than our calculations suggest. We note that the measured spectra shown in Figs. 4–7 are raw spectra, uncorrected for the transmission function of the monochromator, etc., whereas the e-Ar spectrum of Fig. 3 is corrected. Such corrections will affect the line shapes somewhat. Moreover, it is important to keep in one's mind that several different spectral profiles of argon SL bubbles are known that are not in exact agreement; differences of slopes and curvatures are usually discernible. In other words, one can hardly expect perfect agreement in a comparison as attempted here. We also mention that in the calculation of the line shapes, a Boltzmann distribution of the electron energies was assumed. More realistic choices (if they could be made) would certainly affect the resulting line shapes. In any case, at least for the heavier rare gases, the agreement of calculated and observed spectral profiles seems remarkable. At the same time, the somewhat different slopes seen for the lighter rare gases do not seem to be necessarily inconsistent with theory. We will discuss further examples of the variations of measured profiles below.

Next we turn our attention to the comparison of absolute

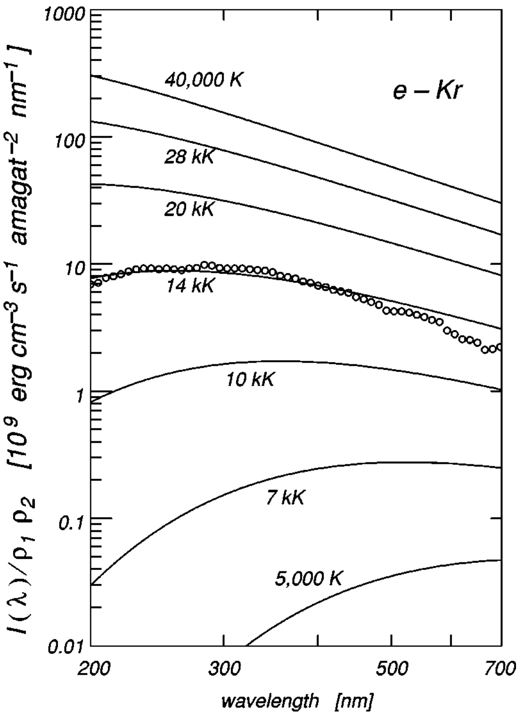

FIG. 4. The e-Kr emission spectra in the spectral window of liquid water, as a function of wavelength, for seven temperatures from 5000 to 40 000 K. A measured SL spectral profile of krypton bubbles is also shown (circles; from Ref. [4]).

intensities. We know that each SL light flash emits roughly 10^5-10^6 photons in the spectral window of water [4]. For the sake of comparison, we integrate our data (Figs. 3–7) over wavelength, after dividing by the energy of a photon, $\hbar\omega$. Figure 8 shows the number of photons emitted per second, per unit volume, and under stationary conditions for the rare gases. For the comparison of calculated and observed photon yields, these integrals must be multiplied by four fac-

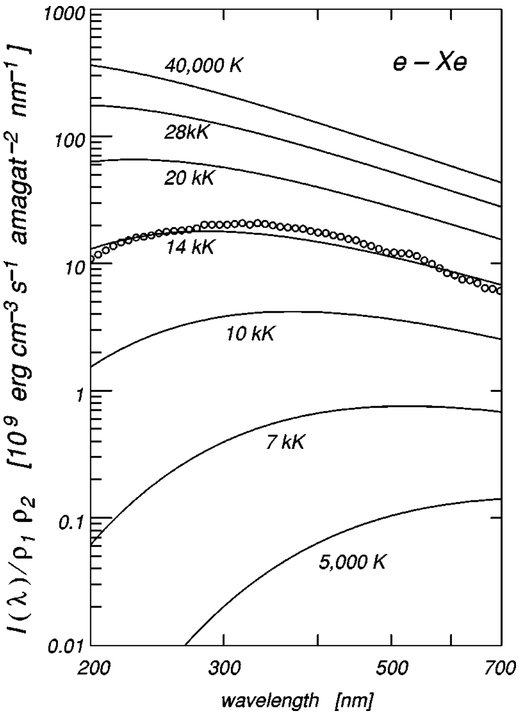

FIG. 5. The e-Xe emission spectra in the spectral window of liquid water, as a function of wavelength, for seven temperatures from 5000 to 40 000 K. A measured SL spectral profile of xenon bubbles is also shown (dots; from Ref. [4]).

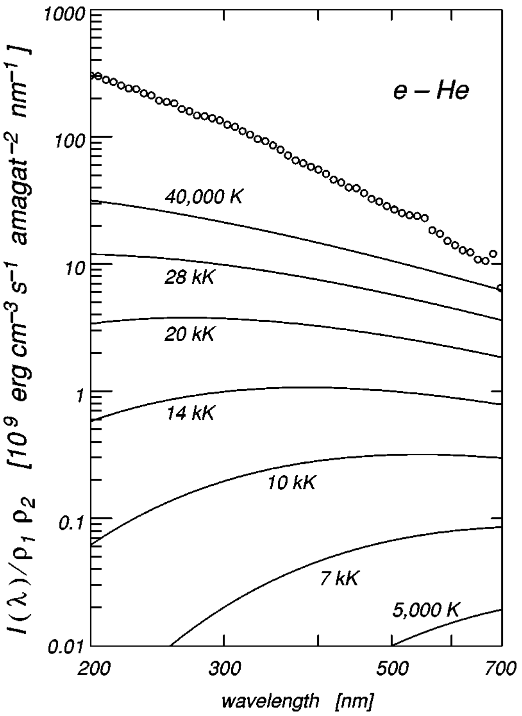

FIG. 6. The e-He emission spectra in the spectral window of liquid water, as a function of wavelength, for seven temperatures from 5000 to 40 000 K. A measured SL spectral profile of helium bubbles is also shown (circles; from Ref. [4]).

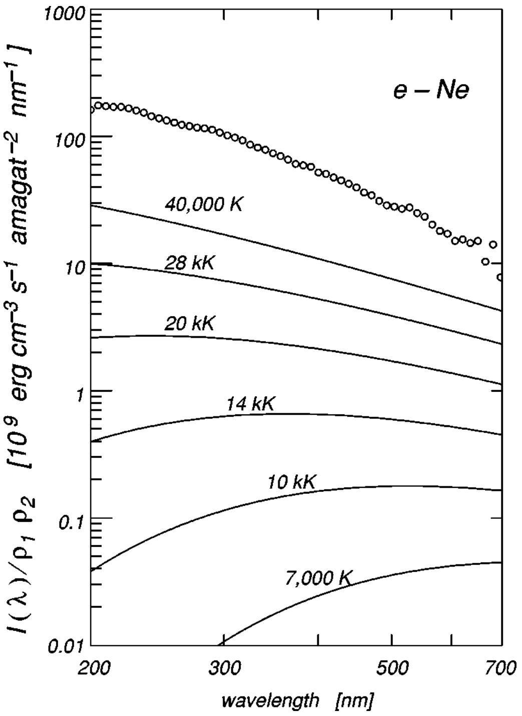

FIG. 7. The e-Ne emission spectra in the spectral window of liquid water, as a function of wavelength, for seven temperatures from 5000 to 40 000 K. A measured SL spectral profile of neon bubbles is also shown (circles; from Ref. [4]).

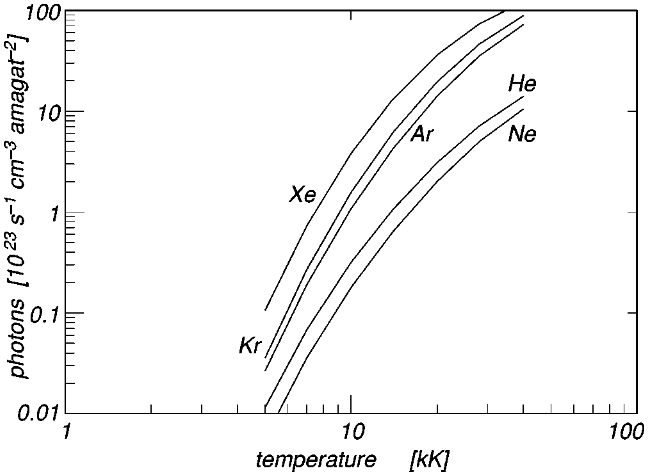

FIG. 8. Number of photons emitted per second, per cm^3, per amagat^2, by collisional electron–rare-gas atom pairs, as a function of wavelength, for temperatures from 5000 to 40 000 K, in the spectral window of liquid water.

tors, namely, (i) the duration τ of an SL flash, (ii) the volume V of the source, (iii) the density ϱ_1 of argon, and (iv) the density ϱ_2 of the electrons. The duration τ is not well known. Previously, it has been argued that τ must be ''much smaller than 50 ps,'' but recently values around 100 ps have been demonstrated [31]. The volume V must be estimated from the smallest bubble radius $R_{\min}$, which is thought to amount to roughly 0.5 μm. The density of argon at maximum compression amounts to roughly 500 amagat (i.e., near the liquid state density) and the electron density should be a small fraction thereof; see Ref. [4] for details concerning the first three factors. The exact value of the product of τ, V, ϱ_1, and ϱ_2 is not known, but for electron densities of roughly 1% of the neutral density we may assume it to amount to roughly

$$\tau V \varrho_1 \varrho_2 \approx 10^{-10}\ \mathrm{s} \times 10^{-12}\ \mathrm{cm}^3 \times 2500\ \mathrm{amagat}^2,$$

or 0.25×10^{-18} s cm^3 amagat^2. Applying such a factor to the coordinate values (Fig. 8), we obtain the right photon numbers per SL flash. For example, for argon bubbles, for temperatures around 20 000 K, we thus expect roughly 0.5×10^6 photons per flash.

Figure 8 also shows that theory suggests a 50% greater intensity for helium than for neon. This is in (quantitative) agreement with the measurements [4] if we may assume that SL in helium and neon generates comparable temperatures. SL of all the other rare gases glows brighter than that of helium, again in semiquantitative agreement with the computations shown in Fig. 8.

IV. DISCUSSION AND CONCLUSIONS

We have seen that reliable, computed bremsstrahlung spectra of electrons colliding with neutral atoms at temperatures around 20 000 K are similar to the observed SL profiles of the rare gas bubbles—certainly for the heavier rare gases if not for all. Moreover, at such temperatures, the computed intensities are consistent with the observed emission of roughly 10^5–10^6 photons per flash reported in the literature. We conclude that the free-free emission mechanism involving electrons and neutral atoms is most likely significant for SL of the rare gases. Moreover, the theory of bremsstrahlung from collisions of electrons with neutral atoms predicts correctly that SL bubbles of helium glow $\approx$50% brighter than those of neon, while all the other, more massive gases glow brighter than helium, in semiquantitative agreement with the theory presented above.

An essential prerequisite for the above considerations is the weakly ionized SL environment. Under stationary (non-SL) conditions, at temperatures around 20 000 K and near liquid state densities, the degree of ionization would be doubtlessly fairly high: a significant fraction of the atoms would be ionized and the emission spectra significantly modified relative to the figures shown above, for a number of reasons. However, these facts need not necessarily invalidate what we have argued above, because the lifetime of the SL environment is measured in picoseconds, that is, just a few times the duration of the neutral-neutral collisions. The cross section for ionization by neutral-neutral collisions is very small, so that it takes many such collisions to generate electrons (e.g., the *first* electrons) if energies are distributed according to Maxwell-Boltzmann statistics. In the shock model of SL [3], the distribution of neutral speeds would be even narrower, and ionization even less likely at temperatures well below the ionization energy (2 eV vs 15.7 eV for argon). We feel that because of the presumably short lifetimes of the SL environment, the assumption of a weak ionization may not be unreasonable.

The absorption coefficient (Fig. 2) is also of interest in SL studies. The reciprocal absorption coefficient α is the mean free path for absorption of a photon. Multiplying the data shown in Fig. 2 by the gas density (500 amagat) and electron density (assume 5 amagat, or 1% ionization), in the spectral window of water, we obtain a value of $\alpha \leqslant 250\ \mathrm{cm}^{-1}$, or a mean free path of 40 μm or greater. That is much greater than the size of the SL source ($\approx$0.5 μm). In other words, the electron–neutral-atom source is optically thin. Under such conditions, Planck's blackbody law, which has often been used to obtain temperature estimates of SL, is not applicable to analyses of the observed SL spectra at wavelengths in the spectral window of water. However, at the lowest frequencies shown in Fig. 2, the absorption coefficient is three orders of magnitude greater. At long wavelengths the source is optically much more dense. As a consequence, the specta in the far infrared (if they could be observed) will differ from Fig. 1 as they approach the familiar blackbody shape (if the environment exist long enough so that equilibrium is established).

We note that the bremsstrahlung model of SL, as presented above, is at this point a highly simplified one. We have considered the radiation only from binary collisions of electrons with atoms, but at the densities mentioned one might wonder what many-body effects would contribute to the observable radiation. Moreover, we assume ''pure'' rare gas bubbles, neglecting all the other particles that might exist under SL conditions, e.g., water vapor, gas impurities, ions, and the effects the SL environment may have on the composition (chemical reactions). Furthermore, at densities approaching liquid state densities, the spectroscopy of the rare gases is affected by pressure broadening, Stark broadening (if partially ionized as we assume), line merging, collision-induced emission (by neutral-neutral collisions, etc.), and other more or less well known processes of the dense fluid states. For a final determination of the emission processes of SL, one has to consider all of these processes in terms of discernible contributions to the SL emission—a lengthy project which will perhaps mature in several years. In other words, this work is just a beginning. We have looked at a variety of other emission processes known to exist in the dense state which were seen not to contribute nearly as much as the bremsstrahlung mechanism does. At this point it seems to us that the latter may indeed be a most promising concept in SL research.

Specifically, we have looked at the spectroscopic effects of ion-neutral collisions, e.g., Ar^+-Ar: ions when accelerated in the field of the atom emit much like all charged particles, only at much lower frequencies, so that ion bremsstrahlung emission may safely be ignored here. Ions also polarize atoms during interaction, which gives rise to a quasimolecular, ''collision-induced'' emission, again generally at frequencies in the infrared, unless temperatures are higher than assumed here [32]. Moreover, one can hardly ignore certain impurities, specifically H_2O and perhaps the OH radical, which are strongly polar and also give rise to collision-induced emission (e.g., by polarizing the neutral atom in the electric dipole field of H_2O, and via exchange forces [32]). In the case of molecular impurities, collision-induced bands appear that may very well extend significantly to frequencies in the spectral window of water. At present, we have looked at a number of such spectra, but so far those spectra seem to be either much weaker or strong only at lower frequencies. Supramolecular spectra may, however, be important for SL studies of molecular gases.

If indeed electron–neutral-atom bremsstrahlung is an important SL emission process, one might wonder why the SL of argon bubbles is orders of magnitude brighter than, say SL of nitrogen bubbles. Doubtlessly, theory suggests that electrons interacting with neutral molecules such as N_2 emit about the same spectrum as shown in the above figures if the electron energies are the same and if the same number of atoms is considered. But it is well known that pure nitrogen bubbles glow much dimmer compared to argon bubbles. We think that an explanation for this unshakable fact must focus on the possible differences of the electron temperatures in monatomic and diatomic gases: In molecular gases electrons undergo inelastic collisions, transferring translational energy to the rotovibrational levels and reducing their mean energies in the process. In monatomic gases, on the other hand, such cooling cannot occur. Electron temperatures may therefore be significantly lower and, as a consequence, the emission much dimmer in molecular gases, because emission decreases strongly with decreasing electron temperature (Fig. 8).

In the same way, another striking observation may perhaps be understood: the dependence of the SL emission intensity upon the temperature of the water. It is well known that the vapor pressure of water changes by an order of magnitude when temperatures are varied from 0° to 35 °C, and the intensity of SL bubbles changes along with it. At low temperatures, less water vapor is present in the argon atmosphere, and electrons have higher mean energies (and emit more bremsstrahlung) due to the near absence of inelastic collisions with molecules (H_2O, in this case). Figure 9 compares the SL line shapes of the rare gases, taken near 0 °C (squares), and at room temperature (circles) [4] (with arbitrary intensity scales). We notice that for every rare gas (with the possible exception of helium) the room temperature profiles all seem to peak at longer wavelengths than their freezing point equivalents. According to what we have seen in Figs. 3–7, this again would suggest lower shock temperatures when the water vapor pressure is increased (assuming the shock mechanism itself is not much affected—just the emission). The comparison of the different rare gas spectra suggests that the lighter rare gases are less affected by the amount of water vapor, for an unknown reason. Moreover, electron temperatures seem to decrease with increasing mass of the rare gas atoms.

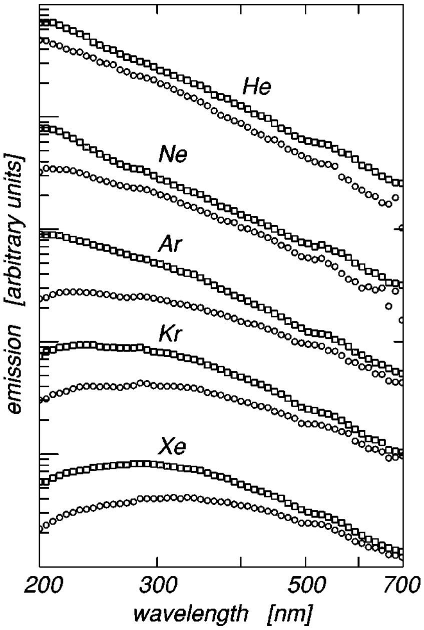

FIG. 9. Comparison of measured spectral profiles of SL rare gas bubbles, taken at near freezing temperatures (squares) and at room temperature (circles).

In conclusion, we mention that previously we have raised a question of whether the glow of sonoluminescencing bubbles of air (or nitrogen, with and without admixtures) may be due to collision-induced emission (CIE) [14]. In the meantime, we have obtained refined calculations and estimates of CIE spectra of a variety of systems that are amenable to *ab initio* calculations. Preliminary results suggest smaller CIE intensities than the (crude) estimate we reported previously for air bubbles [33], but several other systems (e.g., H_2O-N_2 and the ones mentioned above) still may be important and will be considered in future work, especially since the mean electron energies, and thus the bremstrahlung contributions in molecular gases, are not yet well known.

ACKNOWLEDGMENT

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[1] L. Rayleigh, Philos. Mag. **34**, 94 (1917).

[2] T. G. Leighton, *The Acoustic Bubble* (Academic, London, 1994).

[3] C. C. Wu and P. H. Roberts, Proc. R. Soc. London, Ser. A **445**, 323 (1994).

[4] B. P. Barber *et al.*, Phys. Rep. **281**, 65 (1997).

[5] K. S. Suslick, Science **247**, 1439 (1990).

[6] E. B. Flint and K. S. Suslick, Science **253**, 1397 (1991).

[7] R. Hiller, S. J. Putterman, and B. P. Barber, Phys. Rev. Lett. **69**, 1182 (1992).

[8] R. Hiller, K. Weninger, S. J. Putterman, and B. P. Barber, Science **266**, 248 (1994).

[9] R. A. Hiller and B. P. Barber, Sci. Am. (Int. Ed.) **272**, 96 (1995).

[10] D. F. Gaitan *et al.*, Phys. Rev. E **54**, 525 (1996).

[11] K. Weninger *et al.*, J. Phys. Chem. **99**, 14 195 (1995).

[12] H. Frenzel and H. Schultes, Z. Phys. Chem. Abt. B **27**, 421 (1934).

[13] D. Lohse *et al.*, Phys. Rev. Lett. **78**, 1359 (1997).

[14] L. Frommhold and A. A. Atchley, Phys. Rev. Lett. **73**, 2883 (1994).

[15] S. Geltman, J. Quant. Spectrosc. Radiat. Transf. **13**, 601 (1973).

[16] A. Rutscher and S. Pfau, Beitr. Plasmaphys. **8**, 315 (1968).

[17] R. H. Pratt and I. J. Feng, in *Applied Atomic Collision Physics, Vol. 2: Plasmas*, edited by C. F. Barnett and M. F. A. Harrison (Academic, New York, 1984), Chap. 5E, pp. 307–321.

[18] M. Ashkin, Phys. Rev. **141**, 41 (1966).

[19] R. R. Johnston, J. Quant. Spectrosc. Radiat. Transf. **7**, 815 (1967).

[20] C. Yamabe, S. J. Buckman, and A. V. Phelps, Phys. Rev. A **27**, 1345 (1983).

[21] R. L. Taylor and G. Caledonia, J. Quant. Spectrosc. Radiat. Transf. **9**, 657 (1969).

[22] R. T. V. Kung and C. H. Chang, J. Quant. Spectrosc. Radiat. Transf. **16**, 579 (1976).

[23] S. Pfau and A. Rutscher, Z Naturforsch. A **22**, 2129 (1967).

[24] S. Pfau and A. Rutscher, Beitr. Plasmaphys. **8**, 73 (1968).

[25] A. Rutscher and S. Pfau, Physica (Utrecht) **81C**, 395 (1976).

[26] G. M. Weyl and D. Rosen, Phys. Rev. A **31**, 2300 (1985).

[27] J. W. Gallagher, Technical Report No. 16, Joint Institute for Laboratory Astrophysics, University of Colorado, Boulder (unpublished).

[28] V. Kas'yanov and A. Starostin, Zh. Eksp. Teor. Fiz. **48**, 295 (1964) [Sov. Phys. JETP **21**, 193 (1965)].

[29] L. S. Frost and A. V. Phelps, Phys. Rev. A **136**, 1538 (1964).

[30] L. G. H. Huxley and R. W. Crompton, *The Diffusion and Drift of Electrons in Gases* (Wiley, New York, 1974).

[31] B. Gompf *et al.*, Phys. Rev. Lett. **79**, 1405 (1998).

[32] L. Frommhold, *Collision-induced Absorption in Gases* (Cambridge University Press, Cambridge, 1993).

[33] L. Frommhold (unpublished).