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SONOLUMINESCENCE: A NEW ELECTRICAL BREAKDOWN HYPOTHESIS

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We discuss an hypothesis to explain single bubble sonoluminescense as a phenomenon due to electrical breakdown because of large gradients of pressure existing for small radii of the bubble. These large gradients produce large electric fields, flexoelectric effect, that lead to electrical breakdown releasing energies up to 10^{-10}J that is much larger than the light energy released in each cycle. This hypothesis appears to be consistent with several observations made in the sonoluminescense process.

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Sonoluminescence (SL) is the emission of light due to cavitation of gas bubbles in fluids, ordinarily air bubbles in water, during the compression portion of the acoustic cycle. This phenomenon is known more than 60 years [1] but only recently repetitive emission from a single, stable cavitating gas bubble in water has been observed [2]. This made it possible to study the sonoluminescense to considerable detail and prompted discussion of several competing theories to explain the phenomenon (for a recent review and references see, e.g., Ref. [3]). However, the emission mechanism still remains to be understood.

The idea that the sonoluminescense is associated with electrical discharges is almost of the same age as the phenomenon [4,5]. It was considered that the discharge occurs in the bubble or at its surface and several mechanisms of the electric field formation have been proposed [6–8]. However the hypotheses by Frenkel [6] and by Degrois and Baldo [7] have been criticized some time ago [9, 10] and the most recent one [8] seems to be irrelevant to the stationary repetitive luminescense [2]. Indeed, the formation of the electric field is due, within this hypothesis, to detachment of small splinter bubbles with the discharge occuring in the neck region. It is not clear if it is compatible with observation of stationary repetitive luminescense (single emission act per cycle) with simultaneuos control of the bubble radius (no indication of the splinter bubble formation).

In this letter we propose to take into account the electric field created in the environment of the bubble due to gradients of pressure, temperature and dopants concentation. We shall estimate the electric field arising in water due to gradient of pressure (the flexoelectric effect [11]) and shall show that this field is comparable with the breakdown field of water when the radius of the bubble is close to its minimum (a few microns). Thus it is not improbable that the sonoluminescense is a result of an electrical breakdown that as shown below releases energy large enough to explain the observed SL.

The flexoelectric effect has been used already to explain polarization of water by shock waves [12]. Although, it is hardly possible to use the data for the estimation of the relevant coefficients: the estimated time of the shock action [12] is 10^{-12}s while the relaxation time of the polarization in water at 25°C is [13] $0.9 \cdot 10^{-11}$s and the mechanism of the polarization in the shock wave can be quite different from the dipole orientation which dominates for intervals longer than the relaxation time what is our case: the typical time to build the large pressure gradients is 10^{-9}s, much longer than the breakdown time. It should be mentioned as well that there are three flexoelectric effects, bulk static, bulk dynamic and the surface ones [11], but for estimations will consider only the bulk static effect having in mind that all three contribution are, normally, of the same order of magnitude [11].

For the bulk static flexoelectric effect in the spherically symmetrical situation that is expected in the vicinity of an collapsing bubble one has:

$$P = \epsilon_0 \chi E + \alpha \nabla u \tag{1}$$

where P and E are the radial components of the polarization and the electric field, u is the dilation ($u = \Delta\rho/\rho$, ρ is the mass density), χ is the dielectric susceptibility, α is the flexoelectric coefficient. The coefficient α, for ionic solids, is roughly estimated as $\alpha_0 \approx e/a$, where e is the electron charge, a is the interatomic distance. For dipolar substances this is underestimation: at $T \approx 0$ even very small dilatation gradient would provide complete orientation in a system of freely rotating dipoles. To take this into account it seems to be reasonably to estimate α in this case as $\epsilon e/a \equiv \epsilon\alpha_0$ where ϵ is the relative permittivity. Taking into account that in our case $\varepsilon_0 E = -P$

one finds that

$$E = -\frac{\alpha_0}{\varepsilon_0} \nabla u = -\frac{\alpha_0 \beta}{\varepsilon_0} \nabla p \equiv f \nabla p \tag{2}$$

where β is the compressibility of water, p is the pressure. Estimating α as 10^{-10} m and taking into account that $\beta \approx 5 \cdot 10^{-10}\,\mathrm{m}^2/\mathrm{N}$, one finds that $f \approx 10^{-7}\,\mathrm{Vm}^2/\mathrm{N}$. The gradient of pressure ∇p at the surface of the bubble is equal approximately to $\rho\ddot{R}$ where R is the radius of the bubble. Now one can use the data by Barber and Putterman [14] who followed the time dependence of the bubble radius using a light-scattering technique and numerical simulations. For the bubble they studied the maximum acceleration of the bubble radius was about $10^{11}\,\mathrm{m/s}^2$ (according to the numerical integration of the Rayleigh-Plesset equations [15] up to $10^{14}\,\mathrm{m}^2/\mathrm{s}^2$). Then the maximum gradient of pressure is about 10^{14} $\mathrm{N/m}^3$ and the field is about 10^7 V/m (or up to 10^{10}V/m if the results of the numerical simulations are taken seriously). The static breakdown field of water is about 10^7 V/m [16] although, to our knowledge, nothing is known about the breakdown at characteristic time of 10^{-10} - 10^{-11}s in micrometer size regions and the field in these conditions may be well 10^8 - 10^9V/m. However it seems clear that given the bubble accelerations and the pressure gradients the flexoelectric effect is at work and will produce breakdown electric fields. At breakdown fields of 10^8V/m the electric energy released at the breakdown in a 2 μm-size region is about 10^{-10}J. This is two to four orders of magnitude larger than the SL energy released in each cycle.

A breakdown is known to be due to a loss of the stability of more or less homogeneous spatial distribution of current realized at low enough electric fields.

In large systems it leads to a current filament (see, e.g., Ref. [17]). In our case the "distance between the electrodes", i.e. the size of the region of strong fields, is in the micrometer region and the "electrodes" are spherical. Being unaware of the details of mechanism of the electrical breakdown in water we can only speculate about the scenario of such a discharge. It could be well formation of the filament in some part of the spherical region and then rapid propagation of the high conductivity region over all the region. This will lead to a "dipolar component" in the angular distribution of the emitted light. As the breakdown in water is of electronic nature [16] the velocity of the propagation is not necessarily the velocity of sound. One can imagine as well that all the spherical region acquires the high conductivity almost simultaneously if the discharge occurs close enough to the stabity loss point at the current-voltage curve. The meaning of this "almost simultaneously" is still to be understood to make a conclusion that this mechanism explains very short time of the emission [3]. The breakdown may occur as well in the gas inside the bubble. Close to the bubble surface the gradient of pressure in the gas is lower than that in the liquid by the ratio of densities of the liquid and the gas but the breakdown field might be lower as well. Now a scenario with an imploding shock wave in the gas contained within the bubble [18] is considered as the most probable one (see references in Ref.[3] and Ref.[19]). Extremely high gradients of the gas pressure and temperature in the bubble center during the shock collapse are theoretically predicted. Even if the real numbers are somewhat different one cannot exclude that due to the high gradients in the region near the bubble center the breakdown occurs just in this region. Our scepticizm results from the observation that the hydrodynamic treatment of the problem did not include, up to now, the effects of the electric fields arising due to gradients of pressure which can be expected to be important just in the most interesting conditions when the radius of the bubble is close to its minimum value, i.e. when the SL flush takes place. The new hydrodynamic theory, taking into account the flexoelectric effect and, possibly, other non-field mechanism of polarization of the liquid has to be considered.

The work hypothesis described above is consistent with the following points:

(i) the liberated electrical energy is enough to explain the energies released in the SL flushes;

(ii) the short duration of the SL flushes is consitent with the electronic nature of the discharge;

(iii) if the bubble is not perfectly symmetric or the breakdown starts in a region , not at the same time in all the bubble, the emitted light will have a "dipolar component" in the angular distribution of the intensity[20]

(iv) the breakdown mechanism may explain the effects of the noble gas doping[21] by the influence of the concentration gradients which are also produce the polarization and the influence on the breakdown voltage of minute noble gas impurities known since long ago as Penning effect[22]

(v) large magnetic fields should modify the SL conditions because they influence the electronic motion and consequently the breakdown conditions

(vi) with the breakdown model the emission will take place at any radius that satisfy breakdown conditions, not necessarily at the minimum radius.

In conclusion it appears to be that the large accelerations of the bubble radius evidence large enough pressure gradients and electric fields (flexoelectric effect) that will lead to breakdown what is a natural explanation of the SL. Therefore the inclusion of these effects in the hydrodynamic equations is unavoidable. We need to know more about the flexoelectric coefficients and the electric breakdown in the SL conditions but it is at work.

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1. H.Frenzel and H.Schultes, Z.Phys.Chem. **27B**, 421 (1934).
2. D.F.Gaitan, L.A.Crum, In *Frontiers of Nonlinear Acoustics*, Eds. M.Hamilton and D.T.Blackstock, Elsevier, New York 1990, p.459; D.F.Gaitan, L.A.Crum, R.A.Roy and C.C.Church, J. Acoust. Soc. Am. **91**, 3166 (1992).
3. L.A.Crum, Phys. Today **47**, N° 9, 22 (1994).
4. V.N.Levshin and S.N.Rzhevkin, DAN SSSR **16**, 407 (1937).
5. E.N.Harvey, J.Amer.Chem. Soc. **61**, 2392 (1939).
6. Ya.I.Frenkel', Zhur.Fiz.Khim. **14**, 305 (1940). G.L.Natanson, DAN SSSR **59**, 83 (1948).
7. M.Degrois and P.Baldo, Ultrasonics **12**, 25 (1974)
8. M.A.Margulis, Zhur.Fiz.Khim. **59**, 1497 (1985) [Russ.J.Phys.Chem. **59**, 882 (1985)]. Ultrasonics **30**, 152 (1992).
9. M.A.Margulis, Zhur.Fiz.Khim. **55**, 154 (1981) [Russ.J.Phys.Chem. **55**, 81 (1981)].
10. C.M.Sehgal and R.E.Verral, Ultrasonics **20**, 37 (1982).
11. A.K.Tagantsev, Usp.Fiz.Nauk **152**, 423 (1987) [Sov.Phys.Usp. **30**, 588 (1987)].
12. P.Harris, J.Appl.Phys. **36**, 739 (1965). P.Harris and H.-N.Presles, J.Chem.Phys. **77**, 5157 (1982).
13. J.B.Hasted, Prog. Diel. **3**, 103 (1961).
14. B.P.Barber and S.J.Putterman, Phys.Rev.Lett. **69**, 3839 (1992); R.Löfsted, B.P.Barber and S.J.Putterman, Phys. Fluids **A5**, 2911 (1993).
15. N.García, A.Hasmy and A.P.Levanyuk, (to be published).
16. Y.Toriyama and U.Shinohara, Phys.Rev. **51** , 680 (1937).
17. H.Fröhlich, Adv.Phys. **3**, 325 (1961).
18. P.D.Jarman, J.Acoust.Soc.Am. **69**, 1459 (1960).
19. L.Kondić, J.L.Gersten, and Chi Yuan, Phys.Rev.E **52**, 4976 (1995).
20. K.Weninger, S.J.Putterman and B.P.Barber, Phys.Rev. E **54**, 2205 (1996).
21. R.Hiller, K.Wenniger, S.J.Putterman, B.P.Barber, Science **266**, 248 (1994).
22. F.M.Penning and Naturwiss. **15**, 818 (1927); Physica **1**, 1007 (1934).