

Radiation of an electric discharge in a cryogenic liquid

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A study of a high-voltage electrical discharge in a mixture of liquid nitrogen and oxygen at atmospheric pressure revealed a broadband emission spectrum. This spectrum is characterized by the merging of vibrational-rotational bands of the substance's components due to significant broadening due to the interaction of the emitting molecules with those of the surrounding medium. Calculations based on this hypothesis yield good agreement with experimental data.

Keywords: emission spectrum, electrical discharge, line broadening, sonoluminescence.

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In the study of a nanosecond electric discharge in liquid nitrogen, continuous emission spectra were observed (see Fig. 8 in Ref. [1]), qualitatively similar to the spectra usually observed in sonoluminescence [2]: relatively wide spectra with maxima in various fields (in particular, the spectra in liquid nitrogen have two maxima) [1]. The nature of such spectra remains unclear, and their explanation usually boils down to a discussion of bremsstrahlung or recombination radiation, as well as the Planck spectrum of blackbody radiation (see, for example, the discussion in Ref. [1]). It is suggested [3] that such spectra are more characteristic of luminescence, despite the fact that the substances studied in such experiments are not phosphors proper. The origin of the continuous spectra under discussion is presumably explained by the illumination of molecules located on the interfacial surface inside the cavitating liquid [3], which is similar to the glow of organic phosphors, in which the radiation is also caused by individual molecules [4]. Questions remained about which molecules were responsible for the glow and how the observed spectra could be quantified.

The radiation spectrum of a low-current ($\sim 10 \mu\text{A}$) DC electric discharge in a cryogenic liquid is considered in this paper. Liquid nitrogen (saturation temperature at atmospheric pressure 77 K) absorbs oxygen upon contact with air (saturation temperature 90 K), therefore, without taking special measures, we can only talk about cryogenic liquid (liquid nitrogen with the addition of liquid oxygen, hereinafter referred to as LO), and not about liquid nitrogen. In such a liquid, an electric discharge is being investigated on an experimental setup used in Ref. [5] to study an electric discharge in oil. A constant voltage source of up to 30 kV is used; cylindrical tungsten electrodes WC-20 with a diameter of 1 mm are placed in an open thermostat in LO at a distance of 0.5 mm from each other. Boiling did not occur on the elements of the installation, i.e. it is possible to conclude that the system is in a quasi-equilibrium state. LO breakdown occurs at a voltage

of 25 kV, a glow is observed in the discharge region and numerous bubbles with a diameter of $\sim 1 \text{ mm}$ are intensively generated. The radiation spectrum is recorded by a calibrated AvaSpec-2048 spectrometer with a spectral resolution of 2.4 nm from above the liquid surface.

The emission spectra of the discharge in LO are shown in Fig. 1: one — from a discharge located at a depth of 1 cm below the surface of the liquid, the other — from a discharge at a depth of 5 cm. As follows from Fig. 1, the spectra are very similar, i.e., the optical path in a liquid does not affect their shape, and when discussing them, one can ignore the absorption of radiation in a liquid and similar effects. According to Ref. [6], the absorption coefficient of radiation with a frequency of $\sim 10^{15} \text{ Hz}$ in air at an atmospheric pressure of less than 1 cm^{-1} even at a temperature of $2 \cdot 10^4 \text{ K}$, i.e., the absorption of radiation in the plasma inside the bubbles is also insignificant.

The „tails“ of spectra have a power dependence with an exponent of $\sim \lambda^{-2.7}$, as follows from Fig. 2. When

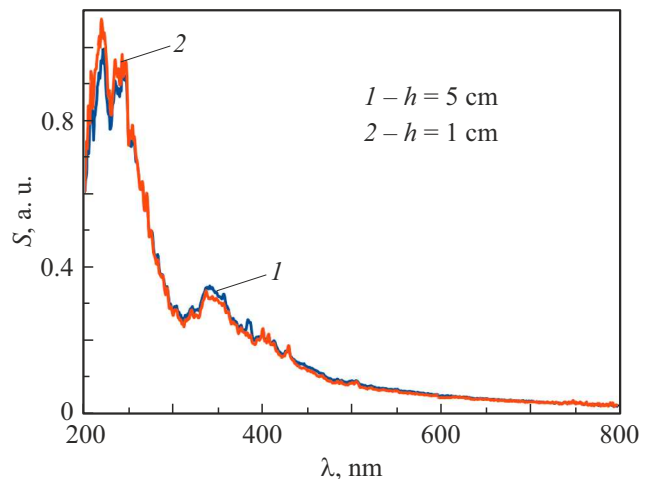


Figure 1. Radiation spectra from a discharge occurring at h different depths inside a cryogenic liquid.

analyzing the spectra, attention should be paid to the fact that the continuous spectra are located in the regions corresponding to the following vibrational-rotational bands:

1) Nitric oxide (NO), transition $A^2\Sigma^+ - X^2\Pi$ — is the so-called γ -series, range $\sim 200\text{--}300\text{ nm}$;

2) molecular nitrogen (N_2), transition $C^3\Pi - B^3\Pi$ — is the so-called second positive series, range $\sim 300\text{--}400\text{ nm}$.

It can be assumed that the observed continuous spectra — are actually strongly overlapping transition lines of the mentioned vibrational-rotational bands. The origin of such a strong broadening is interesting.

In the general case, if the line broadening is due to two factors, the contour of the line is named Voigt, and the frequency distribution is determined by the formula [7]:

$$F_{\Sigma}(\nu) = \int_{-\infty}^{\infty} F_1(\Delta\nu') F_2(\Delta\nu - \Delta\nu') d\nu'. \quad (1)$$

A multiplier c/λ^2 should be added to (1) if a wavelength distribution is considered instead of a frequency distribution.

The broadening of the line due to the interaction of the emitting particle with the surrounding can be described as follows [8,9]:

$$F_c(\nu) = \frac{1}{2\pi} \int_{-\infty}^{\infty} C(\tau) \exp(i2\pi\nu\tau) d\tau, \quad (2)$$

where $C(\tau)$ — is the correlation function. For the first time, an analytical expression for F_c was derived by Lorentz [10], who took an exponential function for $C(\tau)$ and, accordingly, obtained the Cauchy distribution for $F_c(\nu)$. The general expression for the correlation function is written in Ref. [8] if the particles interact according to the law $\sim r^{-p}$:

$$C_{\alpha}(\tau) = \exp\left[-(1 + i \tan \theta)\tau^{\alpha}\right] \quad (3)$$

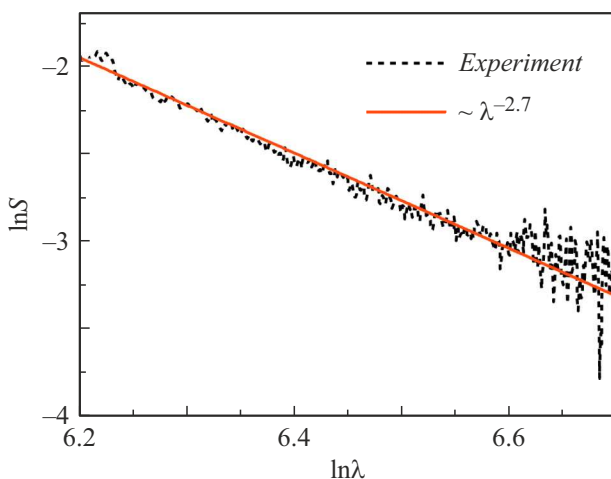


Figure 2. „Tail“ of the radiation spectrum and its approximation by a power function.

(where $\alpha = 3/p$), which essentially corresponds to the characteristic function of the stable distribution [11] over frequencies. In particular, a regular Lorentz contour (Cauchy distribution) is obtained at $\alpha = 1$, while at $\alpha = 2$ a stable distribution corresponds to the Gaussian distribution. Stable distributions have an asymptotic power-law „tail“ $F_c(\nu) \sim \nu^{-1-\alpha}$ at $0 < \alpha < 2$.

Stable distributions are obtained as the limit of the distribution of the sum of independent random variables [11], which is very similar to the principles used to construct the contour of a line widened due to the interaction of a radiating particle with neighboring [8]. In particular, a power-law asymptotic expression for the wings of the line is obtained in Ref. [9], by physical reasoning (see formula 36.44) without reference to stable distributions.

Based on the power-law character of the observed „tail“ distribution (Fig. 2), i.e., knowing the shape of the wings of the line, next we approximate the contour of the line widened by pressure by the function

$$F_c(\nu) \sim \frac{1}{1 + \left| \frac{\nu - \nu_0}{\gamma} \right|^{\beta}}, \quad (4)$$

and calculate the overall contour using formula (1), taking into account (4) and hardware broadening described by a Gaussian function in terms of wavelengths. For the γ -series NO, we take $\beta = 2$ (Lorentz contour), for 2^+ -series N_2 $\beta = 0.7$ (according to the above). The Franck–Condon and Henl–London factors for calculating the intensity of vibrational and rotational transitions are taken from Ref. [12].

The calculation results are shown in Fig. 3 in comparison with the experimental data. A rotational temperature of 80 K is assumed; with such a strong broadening due to particle interaction, this value has little effect on the result. The best match with the experiment can be achieved at an oscillating temperature of 5000 K for NO and 1000 K for N_2 . The parameter γ in (4) is assumed to be $4 \cdot 10^{13}$ Hz for both NO and N_2 . As can be seen from Fig. 3, the match can be considered acceptable. Perhaps the calculation should also take into account the 1^- -series N_2 for a better match in the range of 390–440 nm, however, at this stage, clarifying the details seems unreasonable. For the same reason, we did not consider the line shift.

Thus, the continuous spectrum of the discharge really consists of fused lines of vibrational-rotational bands of LO molecules. The fact that we were working not with pure nitrogen, but with a mixture of nitrogen and oxygen, turned out to be very successful, as it allowed us to compare two groups of vibrational-rotational transitions. It should be noted that the continuous spectra observed in Ref. [1] correspond to the bands of the first and second positive nitrogen series.

The resulting difference in oscillating temperatures does not look strange, since NO molecules are formed in chemical reactions in an electric discharge. The value of the parameter $\beta = 0.7$, which had to be adopted for nitrogen,

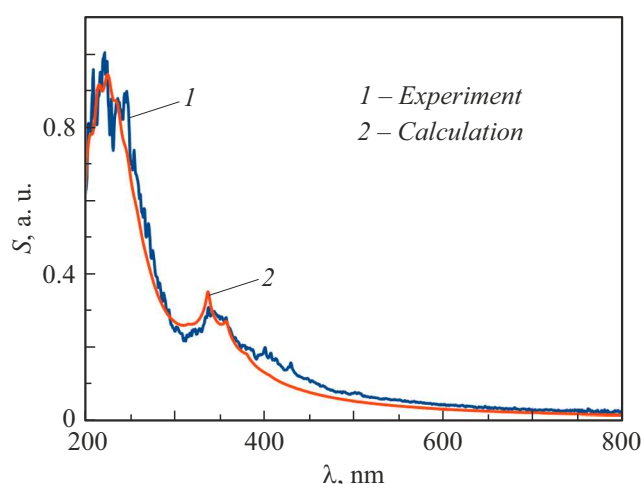


Figure 3. Comparison of the calculated spectrum with the experimental spectrum.

is more difficult to explain. As follows from the above, with such a value of the indicator, the asymptotic behavior (4) does not correspond to any stable distribution. Obviously, the actual contour of the line does not necessarily have to obey such a simple formula as (4), but the behavior of $F(\nu)$ at $\nu \rightarrow \infty$ in a dense environment requires detailed consideration. Taking into account correlations seems to be necessary in the theoretical description of the observed broadening.

From a physical point of view, strong broadening should be caused by the intense interaction of radiating molecules with environmental molecules, which can be observed either in a gas at high pressure or in a liquid. The first option is unlikely to be suitable, since the experiments were conducted at atmospheric pressure. The radiation of molecules from the volume of a liquid can be considered, but it is necessary to explain how excited molecules end up inside the liquid. In addition, in the stationary case, frequent collisions should lead to a Boltzmann distribution with a medium temperature [13], although relaxation of vibrational levels in a dense medium may have its own characteristics in a non-stationary process. At the moment, a compromise option seems reasonable: the source of the radiation under discussion are molecules located on the interface between the bubbles appearing in the discharge and the surrounding liquid. The discharge plasma inside the bubble — the flow of electrons — excites the molecules, and the high frequency of collisions is caused by the surrounding liquid.

The radiation intensity of the 2^+ -series N_2 series depends extremely nonmonotonously on the energy of the exciting electrons (a sharp maximum at the energy of ~ 14 – 15 eV) [14], which should affect the relative brightness of the spectral bands emitted by N_2 molecules from the gas phase and from the liquid surface. At low energy of electrons, the cross-section of their interaction with nitrogen molecules is small, and only in the surface layer of the liquid can we expect noticeable excitation of N_2 molecules due

to the higher density of the medium. However, a similar conclusion can be reached for high-energy electrons: in order to effectively excite N_2 molecules, the electrons first need to lose their energy, which will happen precisely on the surface of the liquid. In both cases, the emission of molecules from the gas phase will be relatively weak. It is difficult to clarify the details of plasma kinetics without a good understanding of the physics of liquid breakdown, and this issue is still debatable [15].

In conclusion, let us assume that the sonoluminescence spectra described in the literature can, most likely, be described in the way discussed above: these can be fused OH hydroxyl bands for water, Swan bands for carbon-containing liquids, etc.

Conflict of interest

The authors declare that they have no conflict of interest.

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