Anomalous luminescence of disperse media during generation in whispering-gallery modes

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A theory is derived for the luminescence of a medium with a condensed disperse phase during the excitation of light in spherical particles which serve as microresonators. It is predicted, for the first time, that there can be a significant intensification of the luminescence of a disperse medium which contains molecules with a population inversion.

1. Recent years have seen increased interest in the physical and chemical properties of heterophase disperse media (see, for example, the review[1] of experimental research on the luminescence of droplets of dye solutions). Our purpose in the present study was to learn about the luminescence of heterophase media under conditions corresponding to the excitation of light in disperse particles.

2. We consider a spherical insulating particle as an open optical resonator. In a spherical resonator, as we know, slowly decaying surface electromagnetic oscillations or "whispering-gallery modes" can exist. Each such mode is characterized by a set p of indices n, k, q, m, where n is a large integer, $k=1$ and $k=0$ for electric and magnetic oscillations, respectively, q is the longitudinal index [the index of the root of the equation $Ai(x)=0$, where Ai is the Airy function], and m is the azimuthal index $(-n \leqslant m \leqslant n)$. The angular frequency ω_p of resonator mode p depends on n, k, and q and is given approximately by $(c/Ma)(n+1/2)$, where c is the velocity of light, M is the relative optical refractive index of the resonator material, and a is the radius of the sphere.[2]

For the quality factor of the resonator, Q, we have derived the expression $Q^{-1} < Q_r^{-1} + Q_s^{-1}$, where Q_r is the radiative resonator quality factor found in Ref. 2, given by

$$Q_r = (n+\frac{1}{2})(\frac{\epsilon}{M})^{2k-1} \frac{(M^2-1)^{1/2}}{2} \exp\{2T_p\},$$

$$T_p \equiv (n+\frac{1}{2})(\eta_p - \tanh\eta_p), \quad \cosh\eta_p \equiv \frac{M^2 a\omega_p}{c(n+\frac{1}{2})};$$

and

$$Q_s = \frac{1}{2\pi}(n+\frac{1}{2})(\frac{\epsilon}{M})^{2k-1}(M^2-1)^{-1}(\lambda/b)^2, \quad \text{where } \lambda = 2\pi c\omega_p^{-1},$$

ϵ is the relative dielectric constant of the sphere material, and b is the amplitude of the deviation of the surface of the particle from an ideal spherical shape ($b \ll \lambda$).

We assume that the particle is a droplet and that b is equal to the mean square amplitude of the thermal capillary oscillations. In this case, Q_r and Q_s are comparable in magnitude, and Q is at a maximum, for a on the order of 30 μm.

3. There can be a generation of light in whispering-gallery modes in a droplet if the wavelength of the light, λ, is equal to $2\pi c/\omega_p$ and if

$$\frac{1}{Q}\omega_p < c\sigma\Delta, \qquad \Delta \equiv \frac{\int dV \Delta N(r) E_p^2}{\int dV \epsilon E_p^2}, \qquad (1)$$

where σ is the cross section for stimulated emission in the droplet, $\Delta N(r)$ is the density of the level population inversion, and E_p is the electric vector of resonator mode p. Lasing has been observed during optical excitation of droplets of a dye solution 20–30 μm in radius.[1]

4. Let us assume that metastable particles in the gaseous phase of a disperse medium have been excited, and a population inversion has arisen between certain electronic levels. The distribution of metastable particles in the gaseous and liquid phases of the disperse medium is found by solving the equations describing the diffusion, excitation, and decay of the molecules. For $a \gtrsim 30\ \mu$m, we have, inside the droplets,

$$\Delta N(r) = yN_g \frac{d_g}{d_l} \frac{L_l}{a} \exp\left\{ \frac{r-a}{L_l} \right\},$$

where y and N_g are the degree of electronic excitation and the concentration of the particles in the gaseous phase of the disperse medium, respectively; d_g and d_l are the diffusion coefficients of the excited particles in the gas and in the liquid; $L_l = \sqrt{d_l \tau_{ql}}$, where τ_{gl} is the lifetime of the metastable molecules in the liquid; and r is the distance from the center of the droplet. If an existing disperse medium is excited, we would have $y = y_g \tau_{qd} \tau_{qg}^{-1}$, where y_g is the degree of electronic excitation of the gas in the absence of the liquid phase, at the same intensity of the excitation of the molecules, $\tau_{qd}^{-1} = \tau_{qg}^{-1} + 4\pi a d_g N_k$, τ_{qg} is the lifetime of the metastable molecules in the gas ($\tau_{qg} \gg \tau_{gl}$), and N_k is the concentration of droplets. If the disperse medium is formed during the rapid introduction of droplets in a previously excited gas, we would have $y = y_0 \exp\{-t/\tau_{qd}\}$, where y_0 is the initial degree of excitation of the gas.

The condition for the excitation of electromagnetic modes (1), can be written as a condition on y: $y > y_t$, where y_t is a quantity on the order of $\pi a^2 (\lambda N_g d_g \tau_{ql} \sigma Q)^{-1}$.

We can find the characteristics of the laser light which are achieved during steady-state lasing. The power of the light emitted by one droplet is

$$P = 4\pi a d_g N_g (y - y_t) \frac{hc}{\lambda}.$$

Under lasing conditions, the quantum yield of the emission of the molecules increases. As a result, the intensity of the luminescence of the disperse medium, I_d, can be significantly higher than I_g, the intensity of the luminescence of a homogeneous gas.

5. As an example we consider the possibility of achieving lasing in droplets of oxygen containing metastable $O_2(^1\Delta)$ molecules. A population inversion exists on the electronic–vibrational transition $O_2(^1\Delta, v'=0 \rightarrow {}^3\Sigma, v''=1)$ ($\lambda = 1.58$ μm) at $y \gtrsim 10^{-3}$ and $T < 300$ K.

We consider a disperse medium containing droplets of oxygen with $a = 30\,\mu$m, at $T = 75$ K, and with a mass fraction of 0.1 of the liquid phase. For such droplets we have $Q = 10^8$, and an emission of light occurs in the droplets if the relative concentration of $O_2(^1\Delta)$ in the gas is $y > y_t = 0.02$.

Gaseous oxygen with $y \lesssim 0.9$ has recently been produced.[3] Lasing can thus be achieved experimentally on transitions of metastable $O_2(^1\Delta)$ molecules. For the power emitted by one droplet under the condition $y \gg y_t$ we have $P = 7y$ (mW) and $I_d/I_g \approx 10^6$.

6. Conclusion. Under certain conditions, the presence of a condensed disperse phase in an excited gas will lead to an anomalous intensification of the luminescence of metastable molecules, as a result of lasing in whispering-gallery modes in the droplets, which serve as microresonators. There is accordingly a new possibility for an efficient conversion of the energy of long-lived, electronically excited molecules into luminous energy.

[1]S.-X. Qian *et al.*, Science **231**, 486 (1986).
[2]L. A. Vaĭnshteĭn, *Open Resonators and Open Waveguides*, Sov. Radio, Moscow, 1966.
[3]N. G. Basov *et al.*, in *Proceedings of the Lebedev Physics Institute*, Vol. 171, 1986, p. 30.

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