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# Effects of Inhomogeneous Broadening on Cooperative Spontaneous Emission of Radiation

E. Ressayre and A. Tallet
*Laboratoire de Photophysique Moléculaire, Université de Paris-Sud, 91405-Orsay, France*
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A theoretical interpretation is given of delayed spontaneous emission of light pulses recently observed in HF gas. It is shown that the large delays can be accounted for by inhomogeneous broadening, which can quench the cooperative emission of radiation to the benefit of incoherent decay.

A recent paper[1] reports the observation of cooperative spontaneous emission of radiation by HF gas. The gas is pumped by an intense pulse from an HF laser, which produces a nearly complete population inversion between two adjacent levels of rotation in the vibrational state $v=1$. The principal feature of these experiments consists in the considerable delay of the light pulse emitted by the molecules after the passage of the exciting pulse. A simulation of experiments[1] by the semiclassical theory agrees with the experimental results. The principal conclusion was that the spontaneous emission of pulses is determined by the superradiant time $\tau_R$, and that the inhomogeneous broadening is unimportant.

In a recent work,[2] we have proposed a symmetrized-master-equation (SME) approach to cooperative spontaneous emission of $\mathfrak{N}$ two-level systems, for large samples and eventually with inhomogeneous broadening. Some numerical applications of our equations predicted large delays in the formation of pulses, depending on various parameters of the problem. These delays were explained as follows: An excited molecule, during its decay, interacts with its own field, and with the electromagnetic field emitted by all other molecules. This latter, which leads to the cooperative emission, is the sum of individual contributions with equal amplitudes but different phases according to the spatial distribution of emitters and inhomogeneous broadening. Because of interference, the cooperative effects can be negligible compared with the decay of independent molecules. But, when the amplitudes of the radiation field emitted by each particle have sufficiently increased, the cooperative emission can prevail over the incoherent emission.

In the experiments on HF gas,[1] the conditions for the validity of the SME model are realized. The first two conditions, $L \ll c\Gamma^{-1}$, $L \ll cT_2^*$ [$L$ is the length of the sample, $\Gamma^{-1}$ the radiative lifetime, and $(T_2^*)^{-1}$ the full width of inhomogeneous broadening], are prescribed by the Markoff approximation. Physically, these inequalities mean that *propagation of light* can be neglected. The second limitation of our model is due to the symmetrization of the master equation for the reduced density operator of the two-level system, $\rho(t)$. When nearly all molecules are excited at time $t=0$, the behavior of the system is described by a closed master equation for the symmetric part $\rho_s$ of $\rho$. Then, the SME corresponds to an interpolation between the cooperative emission process and the incoherent emission process,

$$d\rho_s/dt = (\tau_R\mathfrak{N})^{-1}F^2(t/T_2^*)\Lambda_c\rho_s + [\Gamma - (\tau_R\mathfrak{N})^{-1}F^2(t/T_2^*)]\Lambda_I\rho_s. \quad (1)$$

$\Lambda_c$ is the Liouvillian operator associated with

the cooperative decay, and $\Lambda_I$ that with the incoherent decay. $\tau_R$ is defined by

$$\tau_R^{-1} = \mathfrak{N} \int d^3k\, I_0(k) |e^{i\vec{k}\cdot\vec{r}}|_{\mathrm{av}}^2, \tag{2}$$

where $I_0(k)$ is the radiation rate per unit solid angle for a single molecule; for large samples, $(\tau_R \mathfrak{N})^{-1}$ is much smaller than $\Gamma$. The function $F(t/T_2^*)$ is the Fourier transform of the inhomogeneously broadened profile $g(\Delta\omega)$. In the experiments on HF, $(T_2^*)^{-1}$ is defined by the nutation frequency of the exciting pulse, i.e., $(T_2^*)^{-1}$ is proportional to the square root of the pump intensity. Then $g(\Delta\omega)$ will be approximated by a Lorentzian shape.[3] So, from Eq. (1), the expectation values $\langle R_3 \rangle$ and $\langle R^+ R^- \rangle$ obey the coupled equations

$$d\langle R_3\rangle/dt = -\Gamma(\tfrac{1}{2}\mathfrak{N} + \langle R_3\rangle) - (\tau_R \mathfrak{N})^{-1} \times \exp(-t/T_2^*)\langle R^+ R^-\rangle, \tag{3a}$$

$$d\langle R^+ R^-\rangle/dt = -\Gamma\langle R^+ R^-\rangle + 2(\tau_R \mathfrak{N})^{-1} \times \exp(-t/T_2^*)\langle R^+ R^-\rangle\langle R_3\rangle. \tag{3b}$$

In Eq. (3b), the quantum correlations beyond the second order have been neglected; the product $\langle R^+ R^-\rangle\langle R_3\rangle$ is written in place of $\langle R^+ R_3 R^-\rangle$. The intensity radiated into free space is proportional to the derivative of molecular energy.

Equations (3a) and (3b) have been numerically integrated assuming that $\mathfrak{N}$ molecules are excited at $t=0$. For a rotational transition, the transition moment[4] is defined by

$$\mu_{J+1,J}^2 = \mu_e^2 (J+1)/(2J+3),$$

where the electric moment $\mu_e$ of HF is equal to 1.91 D. Then for the transition $J = 3$-2, at 84 $\mu$m, one has $\Gamma = 0.76\ \mathrm{sec}^{-1}$. For $T_2^* = 220$ nsec, we report the radiated pulses with $\tau_R = 4.7$ nsec (Fig. 1) and $\tau_R = 10$ nsec (Fig. 2). The theoretical curves (solid lines) are drawn by fitting their maxima to the experimental ones (dashed lines). We can see that the SME model reproduces well the experiments, though the theory does not take into account losses effects etc. The theoretical delays

$$\tau_{D_1}^{\mathrm{theor}} = 275 \text{ nsec}, \quad \tau_{D_2}^{\mathrm{theor}} = 680 \text{ nsec},$$

have to be compared with the experimental ones,

$$\tau_{D_1}^{\mathrm{expt}} = 600 \text{ nsec}, \quad \tau_{D_2}^{\mathrm{expt}} = 1500 \text{ nsec}.$$

The absolute difference between numerical results and experimental measures is not surprising, since the origins of times are different; notice only that the ratio $\tau_{D_1}^{\mathrm{theor}}/\tau_{D_2}^{\mathrm{theor}}$ is equal to the experimental one. The observed ringing[5] (Fig. 1) cannot be reproduced by our model since, in this master-equation approach, the pulses emitted by the molecules cannot react on them.

We had shown[2] that without inhomogeneous broadening ($T_2^* = \infty$) large delays are expected only if $(\Gamma\tau_R)^{-1} \ll \ln 2\mathfrak{N}$. But in these experiments the inverse inequality is satisfied ($\mathfrak{N} \simeq 10^{15}$–$10^{16}$), and the effects of terms proportional to $\Gamma$ in (3a) and (3b) would be negligible for $T_2^* = \infty$. We find again the classical behavior[6] for $\langle R_3\rangle$ and $\langle R^+ R^-\rangle$, and the maximum intensity appears at $\tau_D^{\infty} = \tau_R \ln \mathfrak{N}$. Then we can immediately conclude that in the present study delays larger than $\tau_D^{\infty}$ are caused by inhomogeneous broadening. The effect of $T_2^*$ on $\tau_D$ can be studied with the help of Eqs. (3a) and (3b). First, when the inequality

$$T_2^*/\tau_R \ll \ln \mathfrak{N} \tag{4}$$

is satisfied, with $T_2^* \ll \Gamma^{-1}$, the radiated pulse intensity

$$I \propto \exp(-t/T_2^*)\langle R^+ R^-\rangle \tag{5}$$

has its maximum for

$$\tau_D = T_2^* \ln(T_2^*/\tau_R), \tag{6}$$

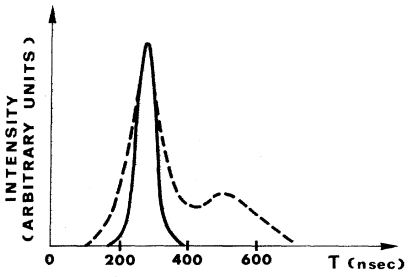


FIG. 1. Intensity of radiation with $\tau_R = 4.7$ nsec and $T_2^* = 220$ nsec. Solid line corresponds the theoretical curve and dashed line to the experimental one taken from Ref. 1.

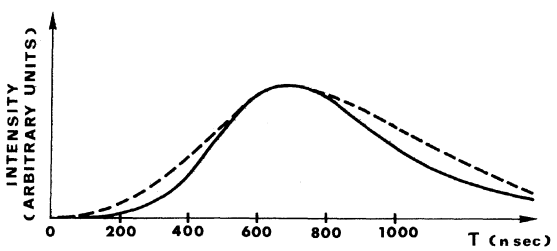


FIG. 2. Intensity of radiation with $\tau_R = 10$ nsec and $T_2 = 220$ nsec. Solid line corresponds to the theoretical curve and dashed line to the experimental one taken from Ref. 1.

with

$$\langle R_3 + \tfrac{1}{2}\mathfrak{N}\rangle = \mathfrak{N} e^{-\Gamma t}. \tag{7}$$

These solutions correspond to the experiment with $\tau_R = 10$ nsec and $T_2^* = 220$ nsec (Fig. 2). The solutions (6) and (7), which agree with the numerical results, show that the emitted pulse (Fig. 2) cannot be categorized as superradiant. Indeed the energy of the system is still near its maximum during the emission of the pulse. This result can explain the absence of ringing at the edge of the observed pulse since the molecular system is nearly saturated. The opposite situation corresponds to

$$T_2^*/\tau_R \gg \ln\mathfrak{N}, \tag{8}$$

which gives the restrictive condition for appearance of superradient emission. In this case, the solutions of Eqs. (3a) and (3b) are

$$\langle R_3\rangle = \tfrac{1}{2}\mathfrak{N}\,\mathrm{th}\left\{\frac{T_2^*}{2\tau_R}\left[\exp\left(-\frac{t}{T_2^*}\right) - \exp\left(-\frac{\tau_D}{T_2^*}\right)\right]\right\}, \quad \langle R^+R^-\rangle = \tfrac{1}{4}\mathfrak{N}^2\,\mathrm{sech}^2\left\{\frac{T_2^*}{2\tau_R}\left[\exp\left(-\frac{t}{T_2^*}\right) - \exp\left(-\frac{\tau_D}{T_2^*}\right)\right]\right\}, \tag{9}$$

$$\tau_D = -T_2^* \ln[1 - (\tau_R/T_2^*)\ln\mathfrak{N}].$$

In the limit of large $T_2^*$, we find again the classical delay $\tau_D^{\infty} = \tau_R \ln\mathfrak{N}$. The solutions (9) correspond approximately to the experimental situation illustrated in Fig. 1. In Fig. 3, we report the variation of $\tau_D$ as a function of $\tau_R$ for different $T_2^*$, obtained by numerical integration of Eqs. (3a) and (3b). The behavior of $\tau_D$ agrees with the above analysis. Formally, from Eq. (6), $\tau_D$ will be zero only for $T_2^* \leq \tau_R$. But, for $\tau_R \gtrsim 12$ nsec, the delays are rather artificial since the maxima of the radiated intensities are very near their initial values.

From the above discussion, we can conclude that in the experiments on HF gas, there are no propagation effects,[7] and that delays larger than $\tau_D^{\infty}$ are due to the inhomogeneous broadening. Depending on the ratio $T_2^*/\tau_R$ we can distinguish three behaviors for the emission of radiation: (i) $T_2^*/\tau_R \gg \ln\mathfrak{N}$. The system emits a well-shaped pulse, with a delay principally determined by $\tau_R$. This is the case illustrated in Fig. 1. (ii) $T_2^*/\tau_R \leq 1$. This is the case of independently decaying molecules. The intensity of radiation is a time-decaying exponential. (iii) $1 < T_2^*/\tau_R \ll \ln\mathfrak{N}$. Formation of pulses is possible. However this emission does not possess the characteristics of superradiance, and the delay is principally determined by $T_2^*$.

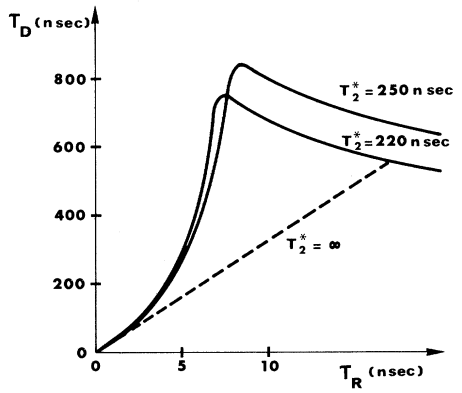


FIG. 3. Theoretical behavior of the delay $\tau_D$ as a function of superradiant time $\tau_R$ for various values of inhomogeneous broadening.

[1]N. Skribanowitz, I. P. Herman, J. C. MacGilliwray, and M. S. Feld, Phys. Rev. Lett. 30, 309 (1973).

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[3]With a driven classical field of constant amplitude and surface $\pi$, the distribution in energy of the molecules $w(\Delta\omega)$ has a Lorentzian shape around the maximum $w(\Delta\omega = 0) = 1$.

[4]C. H. Townes and A. L. Schalow, *Microwave Spectroscopy* (McGraw-Hill, New York, 1955).

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[6]N. E. Rehler and J. H. Eberly, Phys. Rev. A 3, 1735 (1971); R. Bonifacio, P. Schwendimann, and F. Haake, Phys. Rev. A 4, 302 (1971).

[7]Note that the magnitude of $T_2^*/\tau_R$, which is proportional to $\alpha l$, has nothing to do with propagation of light ($\alpha^{-1}$ is the absorption length defined by Beer's law).