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Superradiance spectroscopy: the Te_2 isotopomers

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Superradiance emission in simple systems is a topic of recent theoretical and experimental interest [1 - 4]. It covers the collective radiation damping of an initially inverted assembly of atoms or molecules. The important features of time resolved superradiant emission (delay, intensity and shape) are determined by the number or the density N of cooperating partners. More interesting for spectroscopic applications are the following features : emitted intensity which increases upon N^2 meanwhile halfwidth of bursts of radiation decrease as $1/N$. Hence we report new applications of such a phenomenon to very sensitive spectroscopy concerning hyperfine structure based on analysis of superradiant beats as well as applications to amplifier technology.

As previously mentioned emitted intensity increases with density of partners meanwhile halfwidth of time evolution decreases as $1/\text{density}$. One then has an efficient tool to explore weak emission phenomena such as metastable states or low radiative transitions by forcing such transitions to superradiate. More promising is superradiant spectroscopy which is somewhat connected to quantum beats. Let us recall that when levels involved has no structure or their degeneracy is much smaller than the superfluorescence half-width, time evolution of the emitted light pulse can be represented by a smooth Lorentzian curve (see Fig. 1). However when level structure splittings are of the order of the half-width and upon coherent excitation modulations can be observed on the time-resolved superradiant signal which can be assigned to quantum beats. Yet these beat frequencies are somewhat different from the usual term of quantum beats since in this case it may reflect either initial-level splittings, final-level splittings or a combination of both. In this way we report here observation of beats arising from interference of non-degenerate coherently excited states in the superradiance of rotational transition in $^{125}\text{Te}_2$ vapour and its application to hyperfine structure measurements.

The experimental apparatus is the same as already presented in ref. [3]. Let us briefly recall that we pumped the Te_2 vapour with single light pulses (fwhm = 12 ns) provided by a grazing-incidence dye laser pumped with the third harmonic of a mode-locked Nd-YAG laser. The laser beam was sent onto a T-shaped all-quartz Brewster window cell filled with isotopically pure ^{125}Te (99,8 %), the main cell (length = 7 cm, Fresnel number $\cong 30$) temperature being held at about 900°C. Frequency stability is controlled by a Fabry-Pérot etalon through a beam splitter. Our laser bandwidth is somewhat important (4 GHz). But each mode has a real width of 150 MHz and two successive modes are separated by approximately 600 MHz. Wavelength calibration is achieved

by superimposing the emission spectrum of a Fe-Ne hollow cathode lamp via a pierced mirror. Resolution of the set-up was about 0.003 nm and 1 ns for time dependence. Assignment of spectra was deduced from laser induced fluorescence of $^{130}\text{Te}_2$. Typical modulated signals observed in the forward direction are depicted in Fig. 1.

Fig. 1.

Fig. 2.

As already established [3] main hyperfine contribution arises from excited state accordingly the observed modulation reflects the excited-state splitting as presented in Fig. 2.

Table I presents evolution of beatings frequency which, according to hyperfine interactions dependence varies with the rovibrational parameters v' and J' . We deduced also from our measurements the hyperfine contribution C_E from mixing of $B\ 0_u^+$ state with the other neighbouring electronic states, namely A_{1u} , $A\ 0_u^+$ states which we were not able to derive explicitly from our simple analytic calculation.

TABLE I. Summary of rovibrational evolution of hyperfine contribution C_E relevant of state mixing with $B\ 0_u^+$.

v'	J	Beat frequency (MHz)	C_E (kHz)	$J C_E$ (MHz)
3	90	66	720	64.8
3	108	77	720	83.16
3	118	85	720	84.96
5	134	98	730	97.82

As a conclusion, these results illustrate intermolecular interference effects and point out how they are involved in the appearance of intramolecular ones. Experiments are now in progress to examine in details the evolution of hyperfine contribution with J around perturbed levels in order to give a more precise approach of interactions.

Also among the specific properties of this phenomenon and under certain circumstances, time resolved spectroscopy of superradiant emission exhibits ringing, when the deexcited region of the sample is reexcited by radiation from other parts of the sample and gives rises to bursts of radiation or ringing.

In order to extract observable quantities and to explore the factors which limit ideal behaviour : rotational, vibrational relaxation and Doppler broadening, and the resulting modifications which can occur in the SR emission, we have undertaken a numerical simulation of our experimental data via solutions of the coupled

Maxwell-Bloch equations..Let us recall that in the semi-classical description of a molecular gas interacting with a plane electromagnetic wave propagating along the z direction in the case of slowly varying amplitude leads to a set of differential equations coupling the electromagnetic field E to polarisation density P and population N of a two level quantum molecular (or atomic) system. According to M. S. Feld et al.[2] the evolution of the absorption-emission process can be expressed in the following form :

$$\begin{aligned}\frac{\partial N}{\partial t} &= S - \frac{1}{T_1} (N - N_0) - \frac{1}{2\hbar} (E \cdot P + E \cdot P^*) \\ \frac{\partial P}{\partial t} &= - \left(\frac{1}{T_2} - ikv \right) P + \frac{\langle \mu_z \rangle^2}{\hbar} E N + U \\ \left(\frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right) E &= - \kappa E + 2\pi k P\end{aligned}$$

N represents population difference between the two levels.

N_0 is the initial inverted population.

S represents the optical pumping.

v is the thermal velocity of partners, and k is the wave vector.

U is the contribution of spontaneous emission to P.

T_1 and T_2 represent respectively the longitudinal and transversal relaxation constants. Hence $\frac{1}{T_1}$ includes relaxation processes between the two levels i.e. by rotational, rotational-translational, vibrational and vibrational-translational transfers. On the other hand $\frac{1}{T_2}$ is derived from lifetimes of the two levels as well as homogeneous spectral linewidth.

μ_z is the dipolar transition moment.

κ represents losses.

Integration of these equations has been achieved by a predictor corrector method. In connection with our experimental observations we restricted the emitting medium to a cylindrical sample containing N molecular emitters, the cylindrical axis coinciding with the direction of plane wave excitation and direction $\vec{k}$ of emission. The state of medium is known at any time t therefore one only need determine the field along the cylinder axis, hence giving E (z = l, t) which leads to intensity output of the cell. The onset of superradiance is obtained by initial conditions on the electric field which at z = 0 rely on spontaneous emission only. Our experimental conditions give values from 10^{-5} to 10^{-3} for initial electrical field conditions (in fact it is the reduced field which is correlated to initial Bloch vector tipping angle [2]). Due to the lack of data concerning Te_2 kinetics, the rotational and vibrational relaxation constants which contribute to the $1/T$ term have been derived from its neighbouring molecule, i.e. I_2 [3]. Ignoring Doppler effect the $1/T$ term reduces to a sum $1/\tau_{\text{sup}} + 1/\tau_{\text{inf}}$ where

τ_{sup} and τ_{inf} respectively represent lifetimes of excited and ground state. Lifetimes of rovibrational BO_u^+ states range between 50ns and 90ns and those of ground state XO_g^+ are of the order of 1 μs hence one gets a value of about 0.012 ns⁻¹ for the $1/T$ term. Finally the only parameter left is g , or more physically τ_R , which is the characteristic radiation damping time of the collective emission [2] since $g=1/\tau_R$

Results are displayed in figure 3 with the g values chosen to give the best agreement with the experimental data of figure 2. This tuning on g parameter leads to value of τ_R ranging from 2.4ns to 1.2ns which is in agreement with fitted curves of experimental intensity output [3].

Agreement between the experimental data of figure 2 and the predictions of figure 3 should be considered in the light of the following remarks. If ringing time as well as ringing intensities ratio are well reproduced, the lineshape discrepancy occurs only for ringing side bands. As previously mentioned, our choice of parameters has been restricted to experimental data concerning $^{130}\text{Te}_2$ but we neglected inhomogeneous or Doppler broadening. Subsequent calculations taking into account of this latter effect are now in progress.

References

- [1] R.H. DICKE Phys. Rev. **93**, 99 (1954).
- [2] M.S. FELD and J.C. MacGILLIVRAY in Coherent Non Linear Optics edited by M.S. Feld and V.S. Letokhov (Springer Verlag, Berlin 1980).
- [3] E. BOURSEY, J. MEZIANE, A. TOPOUZKHANIAN IEEE J. Quantum Electron. **29**, 1038 (1993) and Phys.Rev. A **48**,3047 (1993).
- [4] F. HAAKE, M.I. KOLOBOV, C. FABRE, E. GIACOBINO and S. REYNAUD Phys. Rev. Letters **71**, 995 (1993).
- [5] E. BOURSEY, H. ITJI and J. MEZIANE to appear in IEEE J. Quantum Electron. (1994).