PHYSICAL REVIEW A VOLUME 48, NUMBER 4 OCTOBER 1993

# Superfluorescence beats in $^{125}Te_2$ emission

E. Boursey, J. Meziane, and A. Topouzkhanian
*Laboratoire de Spectrométrie Ionique et Moléculaire, Université Claude Bernard-Lyon I,*
*43, boulevard du 11 Novembre 1918, 69622 Villeurbanne Cedex, France*

(Received 26 April 1993)

We report observation of beats in the time-resolved spectra of $^{125}Te_2$ superfluorescence. The results are used to determine hyperfine contributions in the $B\ 0_u^+$ state.

PACS number(s): 33.50.Dq, 42.60.Da, 07.65.−b

We report an observation of superfluorescent beatings in molecular rovibrational spectroscopy. Among the many examples of superfluorescence or superradiance [1–3] the $Te_2$ molecule appears to be, to our knowledge, the best example of molecular superfluorescence [4] since almost all rovibrational states in the 20 000 $cm^{-1}$ range give rise to this effect.

Let us recall that superfluorescence (SF) covers the collective radiation damping process of an initially inverted assembly of partners [1–3] and may be characterized by the two following features: (i) almost all of the energy in the inverted transition is emitted along the excited cylinder, (ii) temporal dependence of the emitted light deviates from the usual exponential decay associated with spontaneous emission and fits a Lorentzian curve [5].

When levels involved in the transition have 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) [5]. However, when level structure splittings are of the order of the half-width and upon coherent excitation [6–8] 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 [6,9]. In this way we report here the observation of beats arising from interference of nondegenerate coherently excited states in the superradiance of rotational transition in $^{125}Te_2$ vapor and its application to hyperfine structure measurements.

The experimental apparatus is the same as already presented in Ref. [4]. Let us briefly recall that we pumped the $Te_2$ vapor with single light pulses [full width at half maximum (FWHM) is 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 is 7 cm, Fresnel number is ≅30) temperature being held at about 900 °C.

Frequency stability is controlled by a Fabry-Pérot étalon through a beam splitter. Our laser bandwidth is

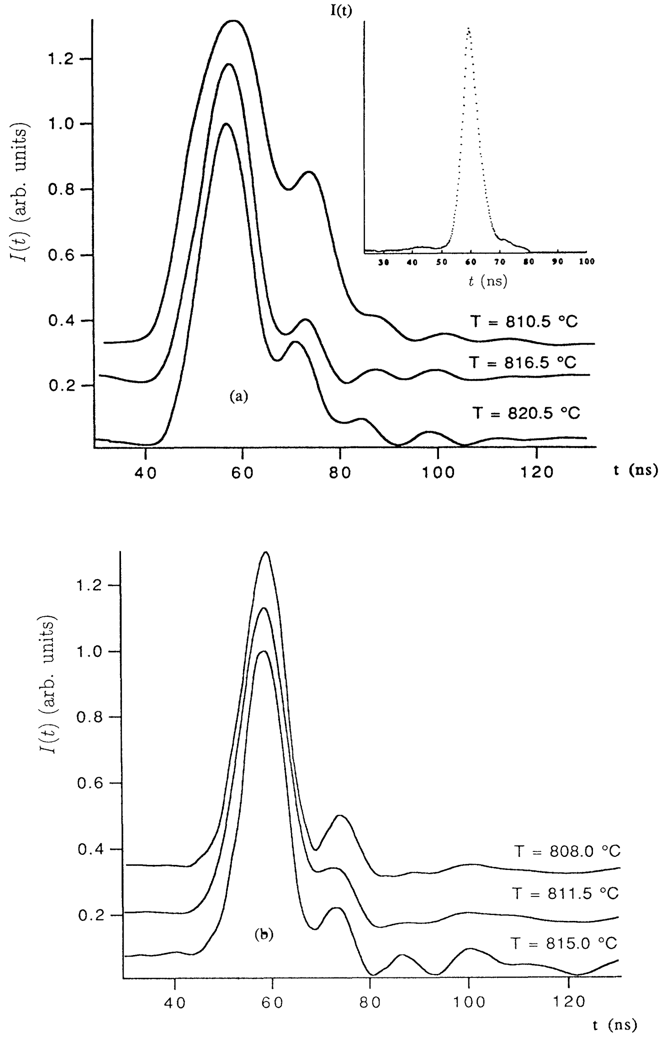

FIG. 1. Representation of evolution of the superfluorescent signal with temperature or density [5], (a) $B\ 0_u^+ \rightarrow X\ ^1\Sigma_g$, $v'=3$, $v''=22$, $J=90$, $\nu_b=65$ MHz, (b) $B\ 0_u^+ \rightarrow X\ ^1\Sigma_g$, $v'=3$, $v''=22$, $J=118$, $\nu_b=85$ MHz. All curves have been arbitrarily shifted vertically and normalized to the same peak intensity. Beats are followed up to 100 ns. Insert: Evolution of the SF signal only for $^{130}Te_2$ [4].

somewhat important (4 GHz). But each mode has a real width of 150 MHz and two successive modes are separated by approximately 600 MHz [10,11].

Wavelength calibration is achieved by superimposing the emission spectrum of an Fe-Ne hollow cathode lamp via a pierced mirror. Resolution of the setup was about 0.003 nm and 1 ns for time dependence. The assignment of spectra was deduced from laser-induced fluorescence of $^{130}\mathrm{Te}_2$ [12–14].

Typical modulated signals observed in the forward direction are depicted in Fig. 1. Among the basic features of SF phenomenon, evolution of peak intensity with temperature or density has been ignored since basically one should expect a dependence upon the square of the number $N$ of emitters. But for molecular partners one has to take into account the evolution of rotational population with temperature, and this somewhat blurs the $N^2$ dependence. It is more effective to derive from data an equivalent relaxation time $\tau_R$ and the start-up delay $\tau_D$ between pump pulse and intensity peak. These two quantities have been extracted by fitting experimental data with an analytical output response derived by Bonifacio and Lugiato [5] which appears to be proportional to $\{\cosh^2[(t-\tau_D)/\tau_R]\}^{-1}$. The evolution of these two parameters with density is displayed in Figs. 2 and 3. Quantum-mechanical or semiclassical Maxwellian treatments yield, respectively, to a dependence of $\tau_D$ on $(1/N)[\ln\sqrt{2\pi N}\,]^2$ [2] or $(1/N)\ln\sqrt{N}$ [5], where $N$ is the density of partners. One shall remark that both models agree with our results since densities are quite high in our experiments and asymptotic values of both models are the same, i.e., $1/N$.

The time-resolved fluorescence intensity depicted in Fig. 1 exhibits a modulated decay which is not the case for $^{130}\mathrm{Te}_2$ [4]. As displayed in Table I, measurement of the frequency of the modulation pattern by Fourier transform leads to a frequency from 65 to 98 MHz depending on rotational and vibrational quanta.

A first-step analysis of these beats can be based on the following remarks: (i) such a modulation has not been observed with $^{130}\mathrm{Te}_2$ [4]; (ii) the modulation pattern is enhanced with increasing temperature, i.e., the number $N$ of partners involved, hence illustrating the effect of the decrease of the relaxation coefficient $\tau_R^{-1}$ in cooperative emission as displayed in Fig. 2; (iii) ringing side bands which are specific to superfluorescence can be ruled out [2,4], since with $^{130}\mathrm{Te}_2$ the ratio of first satellite to main pulse is of the order of $\frac{1}{10}$, which is not the situation as displayed in Fig. 1.

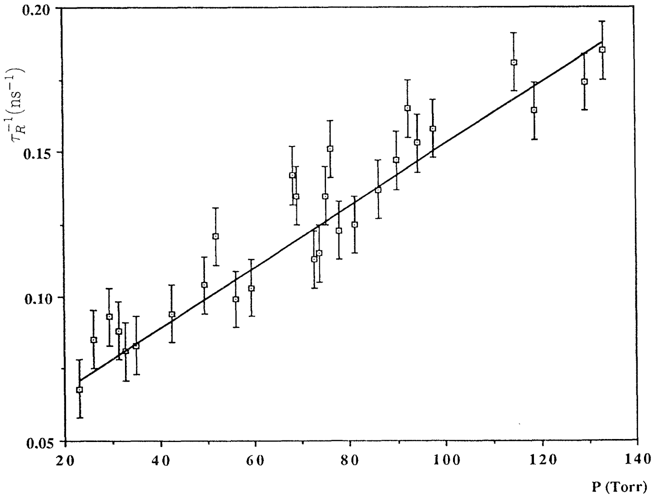

FIG. 2. Evolution of full width half maximum of peak intensity with density.

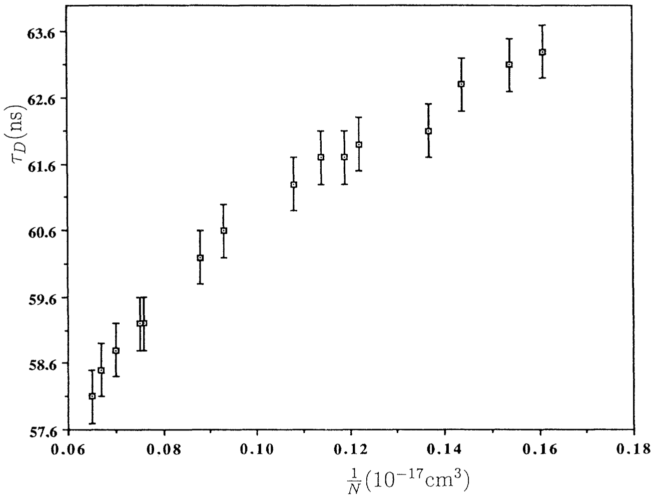

FIG. 3. Evolution of start-up delay $\tau_D$ between the pump pulse and peak intensity with density [4].

One therefore examines the difference between $^{125}\mathrm{Te}_2$ and $^{130}\mathrm{Te}_2$. The latter molecule has a zero nuclear spin, hence Zeeman sublevels are only split in the presence of an external magnetic field as already observed [15]. On the contrary, $^{125}\mathrm{Te}_2$ possesses a hyperfine structure which, according to the $\frac{1}{2}$ nuclear spin of the atom, leads to a splitting ($F=J$, $F=J-1$, and $F=J+1$) of rotational levels in the molecule; hence zero magnetic-field effects can be observed. Modulation of time-resolved signals are then assigned to superradiant beats between hyperfine levels associated to the $B\ 0_u^+ \rightarrow X\ 0_g^+$ transition [9,16–19]. As already mentioned, beat frequencies may be associated with initial-level splitting, final-level splitting, or a combination of both. Hence one is led to obtain direct insight into the hyperfine structure of $^{125}\mathrm{Te}_2$. However, contrary to its neighboring I atom, very little is known about the hyperfine structure of $^{125}\mathrm{Te}$; hence, in support, we have undertaken a rough calculation of the hyperfine degeneracy in the relevant energy levels of $^{125}\mathrm{Te}_2$. As already put forward, $^{125}\mathrm{Te}$ has a $\frac{1}{2}$ spin and

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) | $JC_E$ (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 |

thus an intrinsic magnetic moment. In the molecule these magnetic moments interact with the one associated with molecular rotation, as well as with each other by the magnetic dipole interaction [20–22]. Hence the hyperfine contributions to the Hamiltonian of a diatomic molecule can be expressed in the homonuclear case as [23,24]

$$H_{\mathrm{hfs}}=C\hat{\mathbf{I}}\cdot\hat{\mathbf{J}}+\hat{\mathbf{I}}_1\cdot\hat{\mathbf{d}}(J)\cdot\hat{\mathbf{I}}_2\ ,$$

where the first term represents the spin-orbital moment interaction and the second spin-spin interaction. $\hat{\mathbf{I}}=\hat{\mathbf{I}}_1+\hat{\mathbf{I}}_2$ where $\hat{\mathbf{I}}_1$ and $\hat{\mathbf{I}}_2$ are the angular momentum operators of one of the two nuclei, $C$ is the hyperfine structure constant, $\hat{\mathbf{J}}$ the angular momentum, and $\hat{\mathbf{d}}$ a tensor operator.

Following Bunker and Hanes [23] in the case of $I_2$, the spin-spin coupling term can be expressed for $^{125}$Te$_2$ in the form

$$\hat{\mathbf{I}}_1\cdot\hat{\mathbf{d}}\cdot\hat{\mathbf{I}}_2=1.5d(-1)^{I+F+1}[30(2I+1)(2I'+1)]^{1/2}(2J+1)$$
$$\times\begin{pmatrix} J & 2 & J \\ 0 & 0 & 0 \end{pmatrix}\begin{Bmatrix} F & J & I \\ 2 & I & J \end{Bmatrix}\begin{Bmatrix} \frac{1}{2} & \frac{1}{2} & 1 \\ \frac{1}{2} & \frac{1}{2} & 1 \\ I & I' & 2 \end{Bmatrix}.$$

The $3j,6j$, and $9j$ symbols have been evaluated, according to our observations, for a $J$ value of 120 [24]. The scalar $d$ results from two contributions: direct spin-spin coupling for ground and excited states plus mixing with other electronic states $A\ 0_u^+$ and $B\ 0_u^+$ by nuclear-electron spin coupling for excited state only. The second term suffers from the lack of data concerning Te$_2$ and we made use of a mean value deduced from its neighboring molecule, i.e., $I_2$, which leads to 28 kHz [22,25]. Evaluation of the scalar can be summarized as follows:

$$d_{X\,^1\Sigma_g}(r\sim 2.5\ \text{Å})=4.8\ \text{kHz}\ ,$$

$$d_{B\ 0_u^+}(r\sim 2.8\ \text{Å})=3.4\ \text{kHz}+28\ \text{kHz}\ ,$$

hence leading to a spin-spin coupling contribution of

$$(\hat{\mathbf{I}}_1\cdot\hat{\mathbf{d}}\cdot\hat{\mathbf{I}}_2)_{X\,^1\Sigma_g}=0.81\ \text{kHz}\ ,\quad (\hat{\mathbf{I}}_1\cdot\hat{\mathbf{d}}\cdot\hat{\mathbf{I}}_2)_{B\ 0_u^+}=5.3\ \text{kHz}\ .$$

In either state this contribution appears negligible compared to the observed energy difference of $\sim 80$ MHz. Hence the most important contribution relies on the spin-orbital moment interaction, i.e., the $C\hat{\mathbf{I}}\cdot\hat{\mathbf{J}}$ term. In our case the $\hat{\mathbf{I}}\cdot\hat{\mathbf{J}}$ term can be written [20] as $[F(F+1)-I(I+1)-J(J+1)]/2$ where $J$ is also taken as equal to 120. The hyperfine constant $C$ can be decomposed in two others: (i) a dipole term which is proportional to $\langle 1/r^3\rangle$; (ii) a contribution, in the excited state only, of mixing with other electronic states ($A_u, A\ 0_u^+$) [12] as previously.

The dipolar contribution of the $C\hat{\mathbf{I}}_1\cdot\hat{\mathbf{J}}$ term in the ground state has a value of 7 kHz, hence total hyperfine contribution to ground state $X\,^1\Sigma_g$ is equal to 7.81 kHz. Therefore the main contribution rises from the $B\ 0_u^+$ state; accordingly the observed modulation reflects the excited-state splittings. Moreover, as already described, our laser mode structure allows us to coherently excite neighboring levels only if they are less than 150 MHz apart. This is consistent with the observed modulation frequency of the order of 80 MHz which, as depicted in Fig. 4, may arise from deexcitation of $(J',J'-1)$ or $(J'+1,J')B\ 0_u^+$ levels.

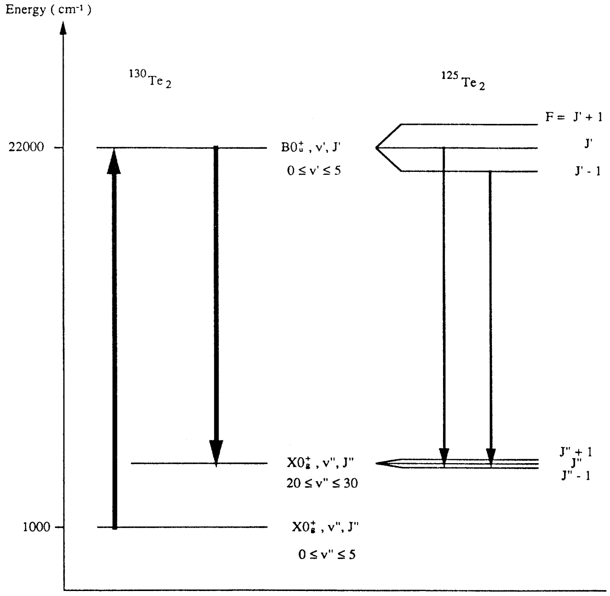

FIG. 4. Relevant energy levels of $^{130}$Te$_2$ and $^{125}$Te$_2$ in zero magnetic field. Hyperfine contribution in ground state of $^{125}$Te$_2$ has been exaggerated with respect to the excited state. The ratio is $10^3$.

Table I presents the evolution of the beating 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 the $B\ 0_u^+$ state with the other neighboring electronic states, namely, $A_{1u}$, $A\ 0_u^+$ [22,23] states which we were not able to derive explicitly from our simple analytic calculation. This result is in agreement with data collected by Townes and Schawlow for similar diatomic molecules [26].

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 detail the evolution of hyperfine contribution with $J$ around perturbed levels in order to give a more precise approach of interactions.

The authors are very indebted to Professor M. Broyer in our Laboratory, and Dr. P. Luc and Dr. C. Pillet from Laboratoire Aimé Cotton for fruitful discussions and comments. The Laboratoire de Spectrométrie Ionique et Moléculaire is "Unité de Recherche Associée du Centre National de la Recherche Scientifique No. 171."

[1] R. H. Dicke, Phys. Rev. **93**, 99 (1954).
[2] M. S. Feld and J. C. Mac Gillivray, in *Coherent Non Linear Optics*, edited by M. S. Feld and V. S. Letokhov (Springer-Verlag, Berlin, 1980).
[3] T. Becker and R. H. Rinkleff, Phys. Rev. A **44**, 1806 (1991).
[4] E. Boursey, J. Meziane, and A. Topouzkhanian, IEEE J. Quantum Electron. **29**, 1038 (1993).
[5] R. Bonifacio and L. A. Lugiato, Phys. Rev. A **11**, 1507 (1975); **12**, 587 (1975).
[6] Q. H. F. Vrehen, H. M. J. Mikspoors, and M. M. Gibbs, Phys. Rev. Lett. **38**, 764 (1977).
[7] J. Marek, J. Phys. B **12**, 229 (1979).
[8] M. Gross, J. M. Raimond, and S. Haroche, Phys. Rev. Lett. **40**, 1711 (1978).
[9] S. Haroche, in *High Resolution Laser Spectroscopy,* edited by K. Shimoda (Springer-Verlag, Berlin, 1976).
[10] M. G. Littman, Appl. Opt. **23**, 4465 (1984).
[11] C. Bordas, P. Labastie, P. F. Brevet, and M. Broyer, Phys. Rev. A **40**, 1696 (1989).
[12] J. Vergès, C. Effantin, O. Babaky, J. d'Incan, S. J. Prosser, and R. F. Barrow, Phys. Scr. **25**, 338 (1982).
[13] E. Boursey, H. Damany, J. d'Incan, C. Effantin, and A. Topouzkhanian, Opt. Commun. **68**, 114 (1988).
[14] J. Cariou and P. Luc, Int. Rep. CNRS Aimé Cotton, 91405 Orsay Cedex, France, 1980 (unpublished).
[15] I. P. Klintsare, M. Ya. Tamanis, and R. S. Ferber, Opt. Spektrosk. **66**, 827 (1989) [Opt. Spectrosc. **66**, 484 (1989)].
[16] W. W. Chow, M. O. Scully, and J. O. Stoner, Phys. Rev. A **11**, 1380 (1975).
[17] M. O. Scully, Phys. Rev. Lett. **55**, 2802 (1985).
[18] M. O. Scully and M. S. Zubairy, Phys. Rev. A **35**, 752 (1987).
[19] J. Bergou and M. Orszag, Phys. Rev. A **38**, 763 (1988).
[20] G. Herzberg, *Molecular Spectra and Molecular Structure. I. Spectra of Diatomic Molecules* (Van Nostrand, Princeton, 1950).
[21] K. F. Freed, J. Chem. Phys. **45**, 4214 (1966).
[22] M. Broyer, thesis, Université de Paris VI, 1977 (unpublished).
[23] P. R. Bunker and G. R. Hanes, Chem. Phys. Lett. **28**, 377 (1974).
[24] B. R. Judd, *Angular Momentum Theory for Diatomic Molecules* (Academic, New York, 1975), and CERN Computer Centre Programm Library.
[25] J. P. Pique, R. Bacis, M. Broyer, S. Churassy, and J. B. Koffend, J. Chem. Phys. **80**, 1390 (1984).
[26] C. H. Townes and A. L. Schawlow, *Microwave Spectroscopy* (McGraw-Hill, New York, 1955).