Eur. Phys. J. B **38**, 395–402 (2004)
DOI: 10.1140/epjb/e2004-00133-x

THE EUROPEAN PHYSICAL JOURNAL B

Saturation of the ODMR effect on singlet exciton induced emission in molecular crystals: Application to crystalline tetracene

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Received 25 August 2002 / Received in final form 22 September 2003
Published online 28 May 2004

Abstract. We have studied the variation of the ODMR effect according to the intensity of a microwave field. We have highlighted a saturation effect of ODMR lines. This effect has been studied as part of the kinetic theory by developing the pilot equation while utilizing for the first time the formalism of a superoperator coupled with a time dependent sinusoidal perturbation. The agreement of the best fit between the experiment and the theory gives the mixing between singlet and quintuplet states, which seems to depend on the direction of the applied magnetic field.

PACS. 71.35.-y Excitons and related phenomena – 32.30.Dx Magnetic resonance spectra – 76.70.Hb Optically detected magnetic resonance (ODMR)

1 Introduction

Optically detected magnetic resonance (ODMR) experiments on delayed fluorescence (DF) due to triplet-triplet exciton annihilation and prompt fluorescence (PF) due to singlet fission into triplet pairs has been extensively studied in molecular [1–6], in charge-transfer [7–11] and in oligomer [12] crystals.

Optically detected magnetic resonance is based on the idea of transferring the detection of a microwave absorption or emission to the optical domain and to take advantage of the concomitant increase in photon energy to enhance the sensitivity of the experiment. The principles of optically detected magnetic-resonance (ODMR) are well known and details can be found in textbooks [13–15].

The detection of the magnetic resonance via the fluorescence, generally termed fluorescence optically-detected magnetic-resonance (F-ODMR), is necessary because the triplet state decays nonradiatively to the ground state and no phosphorescence is observable.

In the molecular crystals the elementary excitations are the hopping neutral Frenkel excitons. The unpaired triplet-exciton population can be generated by a direct excitation in the triplet band and at room temperature the unpaired triplet system can be considered to be in thermal equilibrium with the bath and feeding with equal probability the nine triplet-pair states. One notable exception is that of tetracene crystals in which thermally activated singlet fission can take place and the prompt fluorescence signal can be used to detect microwave modulation with a continuous irradiation in the singlet band at room temperature.

The currently accepted density-matrix theories of triplet-triplet annihilation leading to delayed and prompt fluorescence in molecular crystals are that of Johnson and Merrifield (JM) (kinetic theory) [16] which introduces three phenomenological rate constants [16–18] to describe pair formation, dissociation, and annihilation, and the more elaborate theory of Suna (kinematic theory) [19] which specifically takes into account the triplet motion, notably its dimensionality, to describe the triplet pair formation, and dissociation processes. In most of the studied samples the JM approach was found to be reasonably efficient to account for the observed static-field dependence of the DF and PF signal.

At low temperatures, most of the emission and the microwave-induced resonances are due to heterofusion with trapped excitons and only at room temperature could effects which can be attributed to the free-exciton fusing pairs be observed [20]. At room temperature one deals with free-hopping triplet excitons [21] the theoretical approaches of JM and Suna for nearly two-dimensional materials [19] are, in principle, applicable [22–27].

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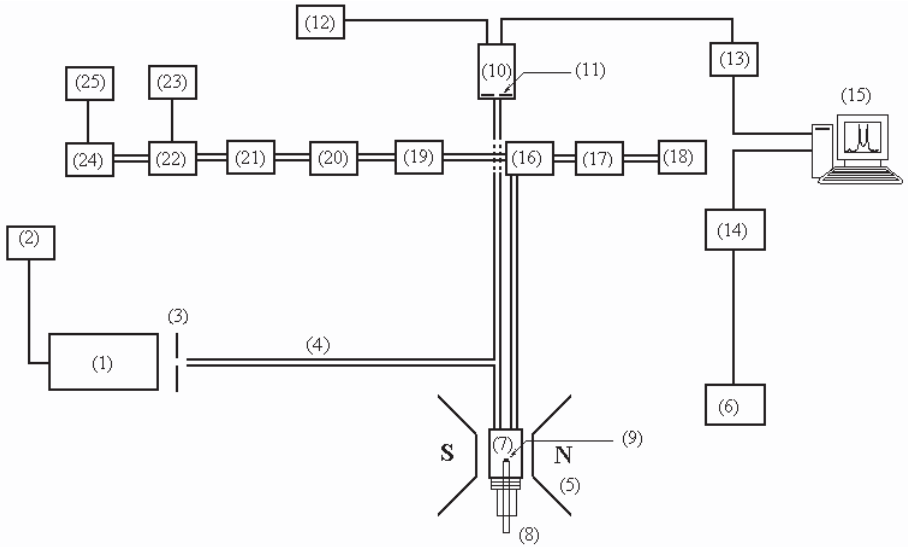


Fig. 1. Experimental set up: (1) Ionised argon laser. (2) Laser Power Supply. (3) Filter F1. (4) Optical fibre. (5) Electromagnet. (6) Electromagnet Power Supply. (7) Sample holder. (8) Quartz tube. (9) Sample. (10) Photomultiplier (PM) . (11) Filter F2. (12) High voltage power supply of the PM . (13) Lockin Analyser . (14) Signal generator. (15) Microcomputer. (16) Circulator. (17) Detector. (18) Oscilloscope. (19) Isolator with ferrite 2. (20) Backward wave oscillator. (21) Isolator with ferrite 1. (22) PIN Diode. (23) Modulator. (24) Gunn Diode. (25) Diode Gunn Power Supply.

At room temperature and for the microwave powers used, the transitions between the unpaired-triplet-exciton Zeeman levels are saturated [28,29] and their contribution to the overall microwave effect on DF involving transitions between pair levels was estimated to be negligible [3,4]. In tetracene, the triplet-pair population was generated by singlet fission into pairs and the microwave-induced resonances were detected via the prompt fluorescence. The microwave effect was found, as expected, to have a sign opposite to that observed for anthracene via DF; that is, one now has a decrease of fluorescence under a microwave field application [5].

ODMR experiments, which simultaneously link smoothness of the optics to the magnetism selectivity were carried out with a fixed microwave power about 4 W. No study, to our knowledge on the effect of the microwave power intensity on the ODMR lines is available.

In this work we propose to undertake an experimental study of the microwave field intensity effect on the evolution of ODMR lines for various directions in the (a, b) plane of tetracene crystals for different thicknesses. In order to interpret these experiments we have proposed in a first theoretical approach a development of the kinetic model by introducing a superoperator formalism.

2 Experimental study

2.1 Experimental set up

Our experiments are performed on crystals of tetracene obtained by sublimation under reduced pressure in a neutral atmosphere and have a thickness of about 50 to 200 μm. The crystalline structure of tetracene is given in reference [5].

The experimental device is represented in Figure 1. We have placed the crystal following the desired plane (a, b), on the top of a quartz guide inside the resonant cavity and we have excited it in its singlet absorption band by the line 4880 Å of an ionized argon laser (*Spectra Physics* 165) through the filter $F1$ (*CS* 5.57).

The luminous excitation has been conveyed by a quartz light guide in order that the laser beam is on the rotation axis of the electromagnet. The fluorescence is recovered by a flexible light guide coming out of the top of the cavity and going towards the photomultiplier (IEM 9558 QB). The fluorescence is analyzed through the filters $F2$ ($Schott$ $BG18 + OG530 + OG570$) which stop the exiting and parasitic photons.

The microwave power (9, 4 GHz) comes from a Gunn diode (30 mW of power) and which is power modulated by a PIN diode (*FMI Model* 16/12) in square signals of 2 kHz is transmitted to the backward wave oscillator (*Top Varian*) which delivers a power of 10 W. The microwave power was modulated in square signals to obtain the envelope of the ODMR signal. The microwave thus amplified, then arrives in the resonant cavity through a circulator. A detector (Fig. 1) allows to check the adaptation of the cavity. Once adapted, the cavity resonates in the mode TE_{011} with a quality factor of $Q = 5 \times 10^3$.

The cavity is placed in the gap of an electromagnet which can turn in the horizontal plane with an angle resolution of $\pm 0.5°$ and whose power supply allows a linear time scan around a central field $H_0 = 3390$ Gauss. The signal which exits from the photomultiplier at an impedance of $Z = 7,5$ kΩ was received by a lockin analyser ($Model$ $SRS830$ DSP) which has as an external reference the PIN diode modulation frequency. The lockin analyser output signal is digitalized using a multimeter ($Keithley$ 195A)

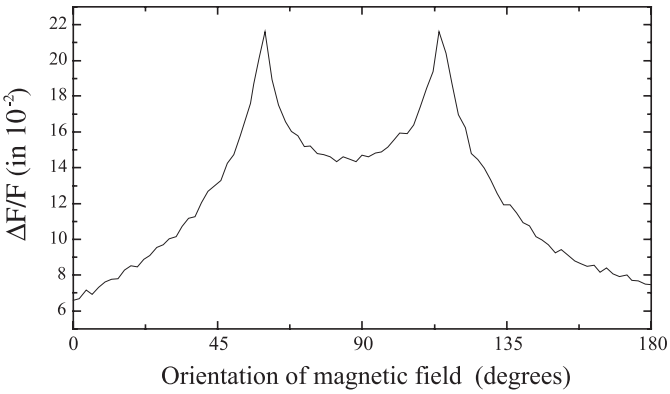


Fig. 2. Anisotropy in the (a, b) plane of the magnetic field dependence at $H = 5 \times 10^3$ Gauss of the prompt fluorescence in crystalline tetracene.

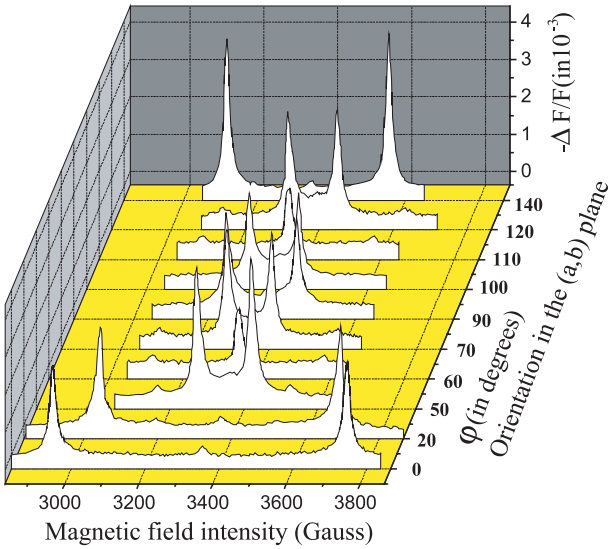


Fig. 3. Experimental ODMR spectra in the (a, b) plane of crystalline tetracene.

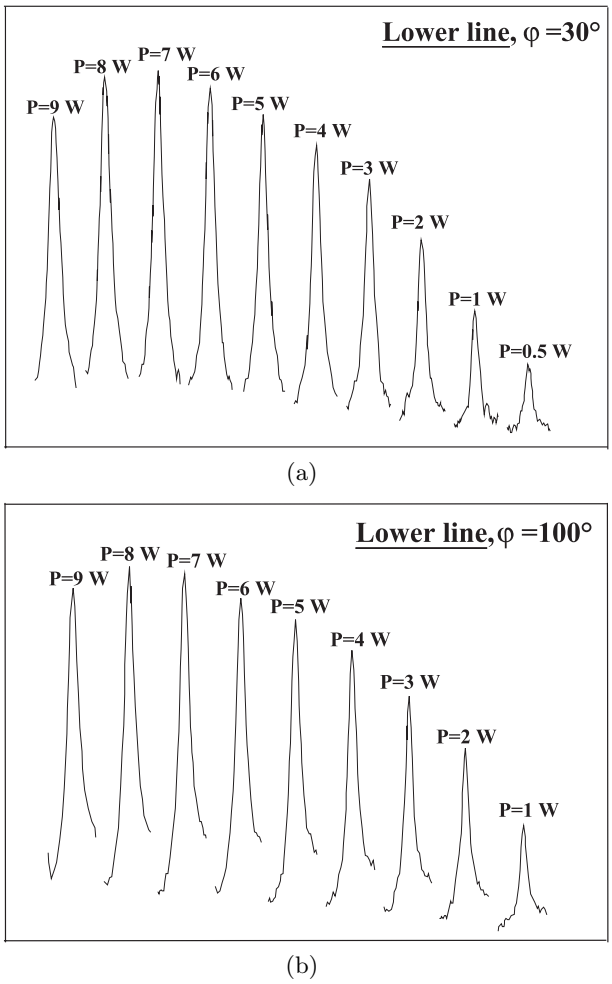


Fig. 4. (a) Evolution of the two lower ODMR lines separately far from the static resonance (for $\varphi = 30°$) as a function of the microwave power for a crystal of thickness 50 μm. (b) Evolution of two lower ODMR lines separately close to the static resonance (for $\varphi = 100°$) as a function of the microwave power for a crystal of thickness 50 μm.

and then transmitted, by an *IEEE* cable, to the microcomputer.

The experimental conditions in which we operated are as follows; the voltage of the photomultiplier is 1200 V, the power of the laser is 300 mW, the sensitivity of the lockin analyser is 10 mV and its response time is 0, 3 s.

The saturation experiments are carried out under the same experimental conditions as those carried out on ODMR experiments. We tried initially to find the minimal intensity of microwave field which gave rise to the ODMR effect. Thereafter, we decreased the attenuation, of the attenuator, in a progressive way until we reached the maximum power while taking care to not damage the crystal, so that all the experiments were carried out under the same experimental conditions.

2.2 Experimental results

In the absence of microwaves, an anisotropy of the prompt fluorescence (PF) in a high static magnetic field is observed in conformity with previous experimental observations [24] as shown in Figure 2. This anisotropy presents two maximum called static resonances located at $\varphi = 59°$ and $\varphi = 112°$ from the crystalline axis $\overrightarrow{a}$.

In the presence of the microwave, one observes a reduction in the fluorescence signal. The intensity of the ODMR lines is about $\frac{\Delta F}{F} = -10^{-3}$. The resonance spectra detected optically (Fig. 3) enabled us to observe a reduction in the fluorescence signal, two intense principal lines for each direction of the magnetic field, a coalescence of the two lines in the neighborhood of the static resonance directions and an anisotropy in the intensities of the ODMR lines, the most intense line is called the upper ODMR line, the other line is called the lower ODMR line.

In Figures 4a and b we have represented the evolution of the ODMR effect for various microwave powers for the lower ODMR lines located far from the static resonance

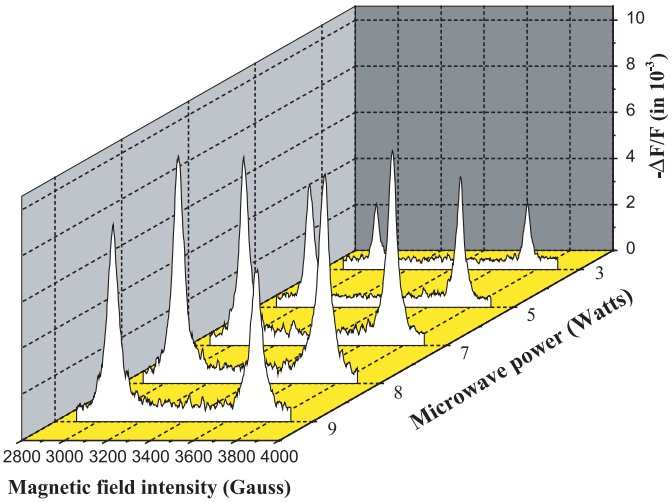


Fig. 5. Evolution of the two ODMR lines together as a function of the microwave power for a crystal of thickness 200 μm.

directions at $\varphi = 30°$ and close to the static resonance directions at $\varphi = 100°$ in a crystal of thickness 50 μm.

In Figure 5 we have represented this same evolution for the two lines (upper and lower ODMR lines) located in the direction at 30° from the crystalline axis $\overrightarrow{a}$ for a crystal of thickness 200 μm.

We notice in both cases a progressive increase in the microwave effect.

We have compared in Figures 6a and b the evolution of the upper and lower ODMR lines peaks according to the microwave power. We notice clearly that these peaks increases initially in a nearly linear way for the low microwave powers until they reach saturation for the high microwave powers, followed by a weak diminution. This diminution is reproducible for all the samples used in this study.

We also notice that this saturation is practically independent of the direction of the static magnetic field and that it is located in the neighborhood of a microwave power of 8 W.

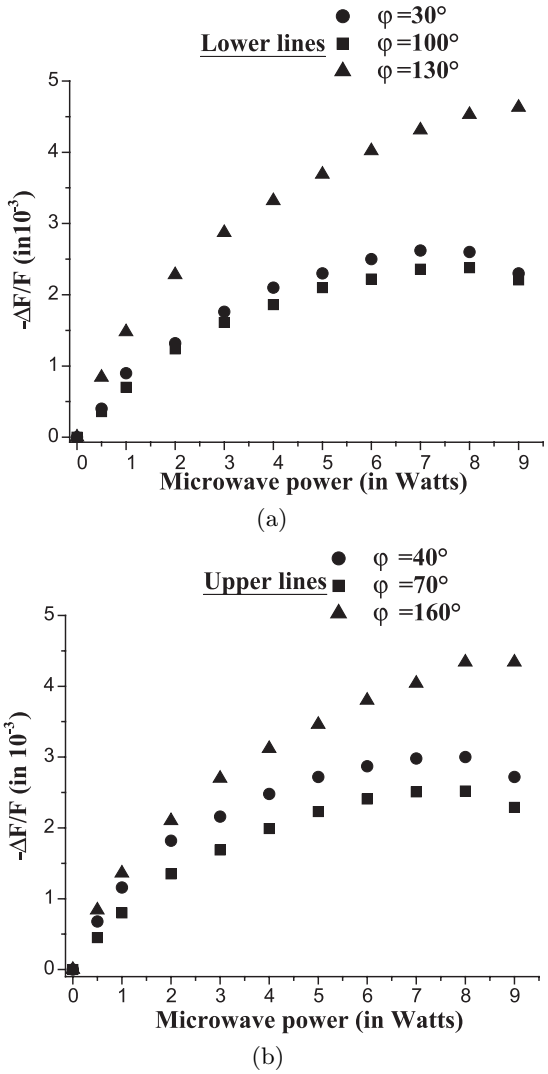


Fig. 6. (a) Evolution of the lower ODMR lines peak as a function of the microwave power for various directions of the magnetic field in the ab plane. (b) Evolution of the upper ODMR lines peak as a function of the microwave power for various directions of the magnetic field in the (a, b) plane.

3 Theoretical study

3.1 Introduction

Small magnetic fields (less than 10 kGauss) modulate the triplet-triplet interaction rate constant γ in some molecular crystals, and therefore the fluorescence intensity, in a characteristic way [30]. To account for these effects, Merrifield proposed that the rate of singlet generation by annihilation of the triplet pair is proportional to the singlet amplitude of the pair, which is determined both by Zeeman interactions and the triplet exciton fine structure [31]. In particular, in high fields, only two pair states have a non-zero singlet amplitude, and a sharp decrease of the annihilation rate occurs when the field direction is such that these two pair states are degenerate. The reaction scheme used is represented in Figure 7, where k_1, k_{-1} and k_2 are respectively the pair formation, dissociation and reaction rate constants leading to a singlet state, k_{-2} corresponds to the opposite reaction rate constant. Generally the two processes of fusion and fission do not coexist for the same material.

This does not meant that (T_1, T_1) is any kind of stable pair, but simply that triplets can be put into two classes, the free and the interacting ones, without any physical assumption about the interaction process. As pairs are short-lived entities, and degeneracy occurs in various cases, interferences among the pair spin states should be considered. For this purpose, Johnson and Merrifield used the full density matrix of the pair spin states [16], from which they obtain an expression for γ, for arbitrary orientation and magnetic field strength; the parameters k_1, k_2, k_{-1} (Fig. 7) and the zero field splitting parameters D^* and E^* of the crystalline triplet may then be computed.

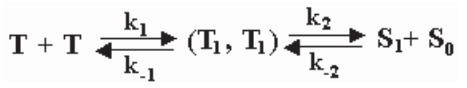


Fig. 7. Kinetic scheme for triplet-triplet pair.

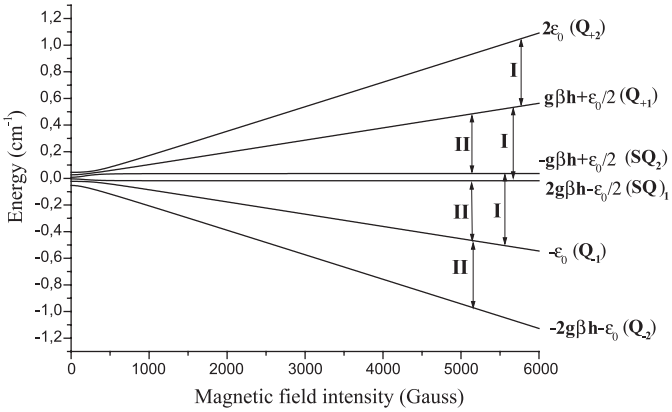


Fig. 8. Possible transitions between the various triplet-triplet pair states.

The kinetics described by Figure 7 gives populations of each state of the triplets exciton pair that are very different from each other; as the lifetime of the pair ($\sim 10^{-9}$ s) is lower than the spin lattice relaxation time ($\sim 10^{-7}$s $\sim 10^{-8}$ s) [32], it must be possible to induce transitions between these states by a power microwave.

The energy differences between the triplet-triplet pair states are about 1 cm^{-1}, it is thus possible to induce transitions between them by exciting the crystal by a microwave field of energy 0.3 cm^{-1}.

Under the microwave effect, the possible transitions are those for which the quantum number m ($\Delta m = \pm 1$) change without variation of the total spin ($\Delta S = 0$). According to this, microwaves could initiate the transitions for the states which have a singlet and a quintuplet character (see Fig. 8).

3.2 Kinetic theory of saturation effect

The process of interaction of two triplet excitons giving a correlated pair is described by the kinetic scheme reported in Figure 7.

The pair-density-matrix evolution can be described by the following:

$$\frac{\partial \rho}{\partial t} = -i\,[\mathcal{H}, \rho] - k_{-1}\rho - \frac{1}{2}k_2\,[P_S, \rho] - \frac{1}{2}k_{SQ}\,[P_{SQ}, \rho] + S, \tag{1}$$

where $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1$, $\mathcal{H}_0$ is the static-field spin Hamiltonian and $\mathcal{H}_1 = g\,\mu_B\,H_1 S_X \cos(\omega t)$ is the perturbation by the microwave field of strength H_1 and frequency ω. P_S being the projection operator on the singlet states S.

In this work we added another kinetic constant k_{SQ} which represents the decorrelation constant of the pair. P_{SQ} is the projection operator on the singlet-quintuplet mixed states. S is the source term.

The solution of (1) can be written in the form:

$$\rho = \rho^{(0)} + \sum_{l=1} \rho'^{\,(l)} \cos(l\omega t) + \rho''^{\,(l)} \sin(l\omega t), \tag{2}$$

where $\rho^{(l)}$ are given by:

$$\rho^{(l)} = \sum_{m=0}^{\infty} H_1^{(l+2m)} \rho^{(l,l+2m)}, \tag{3}$$

which makes it possible to write ρ in the form:

$$\rho = \sum_{m=0} H_1^{(2m)} \rho^{(0,2m)} + \sum_{l,m} H_1^{(l+2m)} \left[Z^{(l,l+2m)} e^{-il\omega t} + \overline{Z}^{(l,l+2m)} e^{il\omega t} \right], \tag{4}$$

and

$$\frac{d\rho}{dt} = \sum_{l,m} H_1^{(2m)} \left[(-il\omega t)\, Z^{(l,l+2m)} e^{-il\omega t} + (il\omega t)\, \overline{Z}^{(l,l+2m)} e^{il\omega t} \right], \tag{5}$$

with

$$\begin{cases} Z^{(l,l+2m)} = \dfrac{1}{2}\left[\rho'^{\,(l,l+2m)} + i\rho''^{\,(l,l+2m)} \right] \\ \overline{Z}^{(l,l+2m)} = \dfrac{1}{2}\left[\rho'^{\,(l,l+2m)} - i\rho''^{\,(l,l+2m)} \right]. \end{cases} \tag{6}$$

To solve equation (5), we use the concept of super-operators defined by:

$$[\mathcal{H}, \rho] = \widehat{\mathcal{H}}\rho = \mathcal{H}\rho\mathcal{I} - \mathcal{I}\rho\mathcal{H}, \tag{7}$$

where $\mathcal{I}$ is the unit operator.

Equation (1) is written then:

$$\frac{d\widehat{\rho}}{dt} + \widehat{A}\widehat{\rho} - \widehat{S} = \alpha \widehat{H}_1 \widehat{S}_X\, \widehat{\rho}\left(e^{i\omega t} + e^{-i\omega t}\right), \tag{8}$$

with:

$$\widehat{A} = -i\,\widehat{\mathcal{H}}_0 - k_{-1}\widehat{\mathcal{I}} - \frac{1}{2}k_2\widehat{P}_S - \frac{1}{2}k_{SQ}\widehat{P}_{SQ}, \tag{9}$$

and:

$$\alpha = -\frac{i}{2}\, g\, \mu_B. \tag{10}$$

$\widehat{\mathcal{H}}_0$, $\widehat{P}_S, \widehat{P}_{SQ}$ and $\widehat{S}_X$ are the superoperators corresponding to the commutator and anticommutator that contain the operators $\mathcal{H}_0, P_S, P_{SQ}$ and S_X in the equation (1). $\widehat{\rho}$ and $\widehat{S}$ indicate the super-vectors corresponding to the vectors ρ and S.

To simplify the expressions we will omit the symbol " ˆ " on the super-operators.

The resolution of the equation (8), gives the following coupled system:

$$\begin{cases} A\rho^{(0,0)} = S, \\ \widetilde{A}Z^{(1,1)} = \alpha S_X \ \rho^{(0,0)}; \quad \text{with} \quad \widetilde{A} = A - i\omega\mathcal{I}\ , \\ A\rho^{(0,2m)} = \alpha S_X \left[Z^{(l,2m-1)} + \overline{Z}^{(l,2m-1)}\right]; \quad m \geq 1, \\ \widetilde{A}Z^{(l,2m+1)} = \alpha S_X \left[\rho^{(0,2m)} + Z^{(2,2m)}\right]; \quad m \geq 1, \\ \widetilde{A}^{(l)}Z^{(l,l)} = \alpha S_X Z^{(l,l)}; \quad \text{with} \quad \widetilde{A}^{(l)} = A - il\omega\mathcal{I} \quad l \geq 2, \\ \widetilde{A}^{(l)}Z^{(l,l+2m)} = \alpha S_X \left[Z^{(l+1,l+2m-1)} + \overline{Z}^{(l-1,l+2m-1)}\right]; \\ \qquad\qquad\qquad\qquad l \geq 2 \text{ and } m \geq 1. \end{cases} \tag{11}$$

3.3 Evaluation of saturation effect

The relative microwave effect of the fluorescence signal is obtained for the fission process from [4]:

$$\frac{\Delta F}{F} = \frac{tr\left[P_S\left(\langle\rho_\mu\rangle - \langle\rho_0\rangle\right)\right]}{tr\left[P_S\,\rho_0\right]}, \tag{12}$$

where $\langle\rho_0\rangle = \rho_0$ is the density matrix in the absence of the microwave field (in the presence of static-field only) and ρ_μ is the density matrix in the presence of the microwave field.

According to equation (2) and (3) we can write:

$$\langle\rho_\mu\rangle = \rho^{(0)} = \rho^{(0,0)} + H_1^2\rho^{(0,2)} + H_1^4\rho^{(0,4)} + H_1^6\rho^{(0,6)} + ..., \tag{13}$$

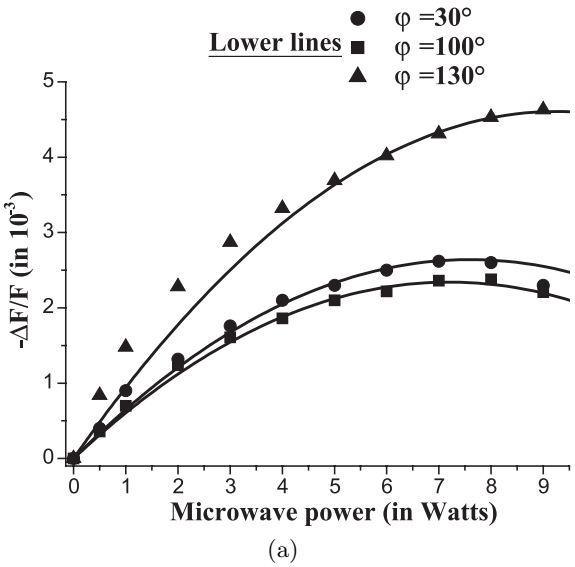
$$\langle\rho_0\rangle = \rho^{(0,0)}, \tag{14}$$

$$\langle\rho_\mu\rangle - \langle\rho_0\rangle = H_1^2\rho^{(0,2)} + H_1^4\rho^{(0,4)} + H_1^6\rho^{(0,6)} + ..., \tag{15}$$

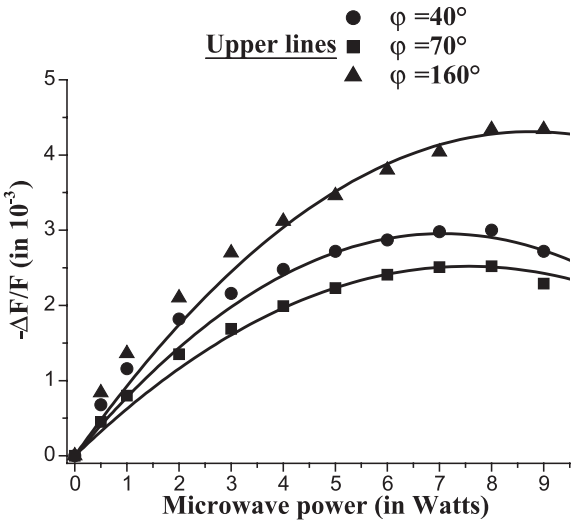
we obtain then in H_1^6:

$$\frac{\Delta F}{F} = \frac{H_1^2 tr\left(P_S\,\rho^{(0,2)}\right) + H_1^4 tr\left(P_S\,\rho^{(0,4)}\right) + H_1^6 tr\left(P_S\,\rho^{(0,6)}\right)}{tr\left(P_S\,\rho^{(0,0)}\right)}. \tag{16}$$

The elements , $\rho^{(0,2)}$, $\rho^{(0,4)}$ and $\rho^{(0,6)}$ are calculated using the system equation (11) which is numerically solved using the functions F07ARF and F07ASF of mathematical library NAG in FORTRAN.



(a)



(b)

Fig. 9. (a) The best fit obtained for the lower ODMR lines. (b) The best fit obtained for the upper ODMR lines.

4 Results and discussion

In order to reproduce the experimental results we used the expression (13) by taking k and k_{-1} equal to the values obtained from the ODMR experiments carried out at average power on our samples. We varied k_{SQ} in order to obtain the best agreement between the experiment and the theoretical model.

In Figures 9a and b we represent the best fit that we obtained for the saturation effect of the ODMR lines for various directions in the (a, b) plane of tetracene.

We notice that saturation is well reproduced (around 8 W) and we obtained a reasonable fit for the evolution effect for the low powers as well as for the high powers.

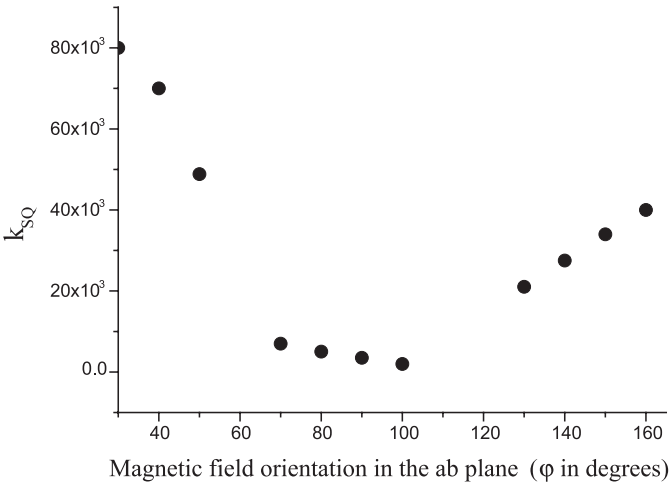


Fig. 10. Evolution of k_{sq} with magnetic field orientation in the (a, b) plane of the tetracene crystal.

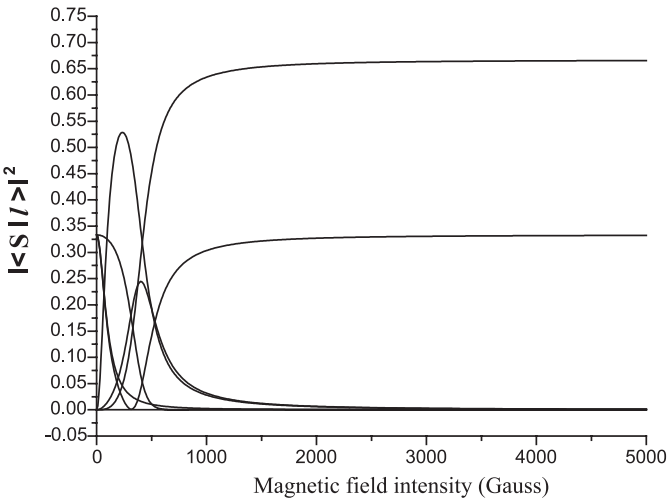


Fig. 11. Variation of the singlet weight with magnetic field intensity.

The kinetic model, which we have used allowed us to explain the saturation effect and to confirm the values of k and k_{-1} obtained using ODMR experiments at intermediate power and for all the directions. It is not the case for k_{SQ} which seems to depend on the direction of the applied magnetic field and which has a minimum near the static resonances (Fig. 10).

Given that, this constant (k_{SQ}) describes the mixing between the singlet and quintuplet states, and since the magnetic field orientation via the singlet weight (Fig. 11) depend on that mixing, this variation seems justified.

5 Conclusion

In this work we have studied for the first time the variation of the ODMR effect according to the microwave power.

The measurement of the ODMR line intensities according to the microwave field intensity for several orientations of the static magnetic field in the (a,b) plane of the crystal shows a saturation effect of ODMR lines. This saturation is practically independent of the direction of the ODMR line and it is located around a microwave power of 8 W.

This effect has been studied as part of the kinetic theory while utilizing a formalism of superoperator coupled with a time dependent sinusoidal perturbation.

We found this best agreement between the experiment and theoretical results while utilizing the constant k_{SQ} which describes the coupling between the singlet state and quintuplet states and which has a negligible effect for the low power microwaves.

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