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FOUR-WAVE MIXING TECHNIQUE AND COHERENCE EFFECT ON ELECTRONIC STATES OF DYE MOLECULE

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Recent papers [1-3] report on the measure of a $T_2$ dephasing time when transient grating experiments are conducted on dye molecules (cresyl-violet) embedded in polymer films. On the other hand it is well known that dye molecules show up a broad continuum of states which is induced through a strong coupling between the electronic and vibrational states of the dye molecule. Therefore such a system can be hardly considered as a two level system and the proposed interpretation remains doubtfull.

We undertook this work in order to bring a more detailed information about this problem. We applied degenerate-four-wave-mixing technique on polymer films doped with cresyl-violet dye at various temperatures and at different excitation wavelengths. We obtain results similar to, but more complete than, those which have been published in Ref.3. We will discuss our results on the basis of the two level model proposed in this reference.

The degenerate 4-wave mixing experiment consists in spatially splitting a laser beam of coherence time $\tau_c$ in two equal intensity beams of wavevector $k_1$ and $k_2$, in delaying one of these two beams by a time $\tau_{12}$. One makes them interfere in the studied sample and measures the total energy diffracted along the 2 $k_1 - k_2$ or 2 $k_2 - k_1$ directions as function of $\tau_{12}$. Both curves exhibit the same width $\Delta$ and a maximum. These two maxima are arranged symmetrically compared to $\tau_{12} = 0$ and are at a distance from each other labelled by definition $\delta t$.

Considering then a two level scheme characterized by a $T_2$ dephasing time, one can theoretically compute $\delta t$ and $\Delta$ as function of $T_2$ and $\tau_c$. For instance, one gets the curve of Fig.1 when one uses the theoretical background of Ref.3.

The value of $T_2$ can be determined by the knowledge of $\tau_c$ and the measure of $\delta t$ by using this simple model. So the experimental measures of $\delta t$ and $\Delta$ allow to plot $\delta t/\Delta$ experimental ratios as function of $T_2/\tau_c$ at different temperatures. We have performed these experiments at different excitation wavelengths. We have got:
Figure 1

Ratio \( \delta t/\Delta \) of the peak time shift to width ratio versus \( T_2/\tau_c \) ratio.

- When exciting the sample on the high energy side or on the top of the absorption band of cresyl-violet, our experimental data can be interpreted with a \( T_2 \) time equal to \( 0 \) (\(<20\) fs).

- When exciting the sample on the low energy side of the absorption band, where the absorption coefficient steeply varies with the wavelength, one gets large variations of \( \delta t \) as function of temperature. The \( \delta t \) experimental values lead to \( T_2 \) values varying from \( 0 \) at high temperature (\( 300 \) K) to \( 200 \) fs at low temperature (<\( 4 \) K). Plotting \( \delta t/\Delta \) experimental values as function of these \( T_2 \) leads to the experimental point of Fig.1. Strong deviation from the theoretical curve is obtained for the experimental points obtained at low temperature. It results then that the two level scheme of Ref. 3 is not able to explain our experimental results.

Moreover we have studied the correlations which exists between the experimental fluctuations on the values of \( \delta t \) and of \( \Delta \). So we can have proved that \( \delta t \) depends linearly on \( \Delta \), which in turn is not predicted by Ref. 3 theory. We have also concluded that \( \Delta \) reflects mostly the coherence time \( \tau_c \) of the laser beam. We give the \( \delta t/\Delta \) experimental values
as function of temperature. This dependence exhibits a critical temperature value ranging around 20–50 K. No interpretation of this critical temperature is yet given.

References