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Using highly efficient Fluorescence excitation spectroscoy of individual pentacene molecular impurities in p-terphenyl crystals, we have observed that some pentacene defects exhibit spontaneous spectral jumps in their resonance frequency at 1.5 K, with a jump rate independent of laser power. In addition, the low-power limiting linewidth for single pentacene absorbers reaches the lifetime limit of 7.8 MHz at 1.5 K.

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by

W. E. Moerner and W. P. Ambrose

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COMMENT ON "SINGLE PENTACENE MOLECULES DETECTED BY FLUORESCENCE EXCITATION IN A *P*-TERPHENYL CRYSTAL"

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ABSTRACT: Using highly efficient fluorescence excitation spectroscopy of individual pentacene molecular impurities in *p*-terphenyl crystals, we have observed that some pentacene defects exhibit spontaneous spectral jumps in their resonance frequency at 1.5 K, with a jump rate independent of laser power. In addition, the low-power limiting linewidth for single pentacene absorbers reaches the lifetime limit of 7.8 MHz at 1.5K.

Comment on "Single Pentacene Molecules Detected by Fluorescence Excitation in a p-Terphenyl Crystal"

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In a recent Letter, Orrit[1] et al. show the fascinating result that the use of fluorescence excitation for single molecule detection (SMD) and spectroscopy in solids can yield excellent signal-to-noise ratio, thus confirming earlier SMD using absorption techniques.[2] We obtain similar results using fluorescence excitation, and wish to comment on two points.[3]

Orrit et al. find anomalously large line widths of 10-15 MHz for single pentacene molecules at 25 mW/cm^2. We find similar widths at similar intensities; however, at lower intensity (1.8 mW/cm^2) the lifetime-limited width[4] of 7.8 ± 0.2 MHz is reached (Fig. 1). We find a measured saturation intensity I_s of 7 ± 3 mW/cm^2 which is an order of magnitude below the three-level I_s calculated from known photophysical parameters[4]. Apparently, while the power-broadening characteristic of single pentacenes is modified, the excited state lifetime is not.

Orrit et al. observe sudden drops and surges in some emission peaks which are interpreted as photo-induced spectral hole-burning of single molecules.[1] While photo-induced changes may occur, we find that two distinct classes of molecules are present: class I, which are stable in time, and class II, which show spontaneous, discontinuous jumps in resonance frequency of 20-60 MHz on a 1-420 s time scale. The occurrence of class II defects is quite common in the wings of the inhomogeneous line (increasing to 40 % at +0.23 nm from line

center), but only class I defects have been observed in a spectral region from 0.003 to 0.01 nm from line center. Fig. 2 shows that for fixed laser frequency, the fluorescence of a class II defect turns on and off in a stochastic fashion as the molecule jumps into and out of resonance. Moreover, the jump rate changes little with laser power (Fig. 2 a,b). We find no evidence that the jumping transitions are laser-driven; the dominant effect appears to be spectral diffusion of class II molecules perhaps because they are coupled to an (unidentified at present) ensemble of two-level systems in the host undergoing phonon-assisted tunneling.

This work was supported in part by the U. S. Office of Naval Research.

REFERENCES

1. M. Orrit and J. Bernard, Phys. Rev. Lett. 65, 2716 (1990).

2. W. E. Moerner and L. Kador, Phys. Rev. Lett. 62, 2535-2538 (1989).

3. W. P. Ambrose and W. E. Moerner, subm. to Nature

4. H. de Vries and D. A. Wiersma, J. Chem. Phys. 69, 897 (1978).

FIGURE CAPTIONS

Figure 1. Fluorescence excitation spectrum for a single pentacene molecule in a sublimed crystal of p-terphenyl at 1.5 K. 0 MHz ≡ 592.407 nm, far into the wings of the O_1 site inhomogeneous line. The solid line is a Lorentzian fit.

Figure 2. Spectral jumps in the resonance frequency of a class II single defect detected with a fixed frequency laser at 592.362 nm. (a) 0.9 nW, (b) 36 nW.

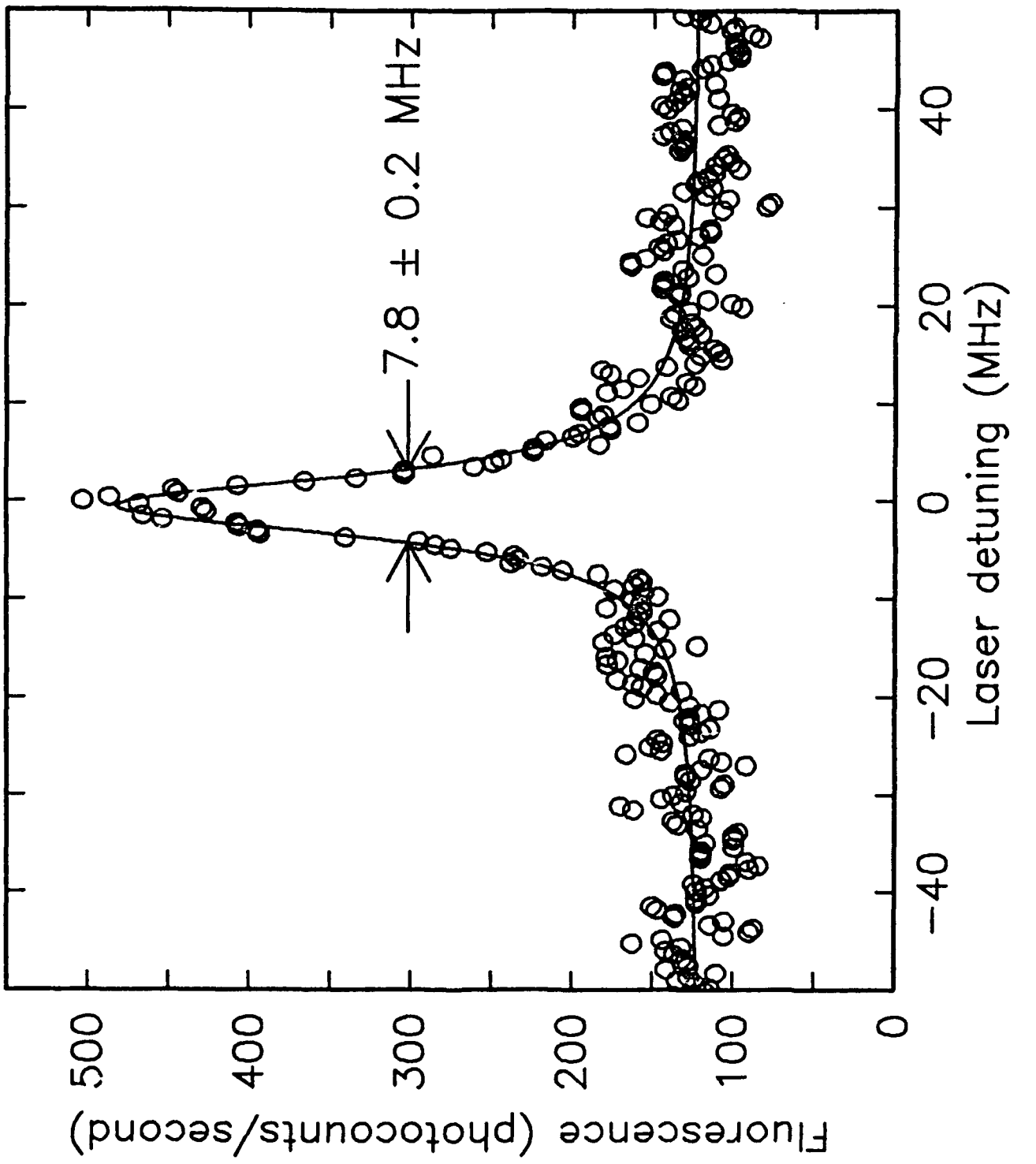
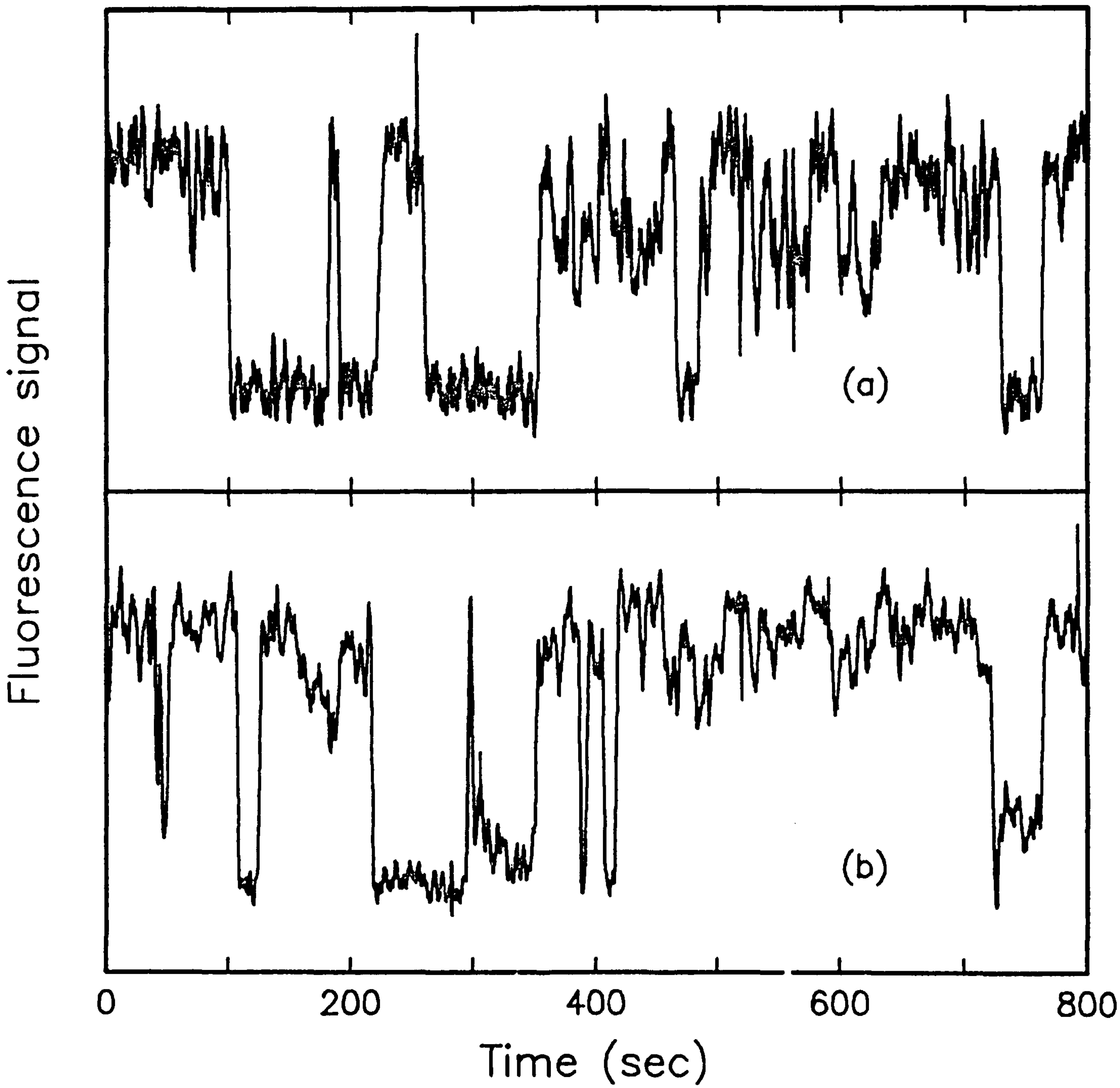

7.8 ± 0.2 MHz
Fluorescence (photocounts/second)
500
400
300
200
100
0
-40
-20
0
20
40
Laser detuning (MHz)

FIGURE 1


Fluorescence signal
(a)
(b)
0
200
400
600
800
Time (sec)

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