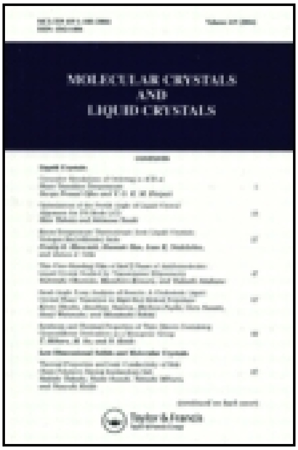

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Jaak Aaviksoo [a] & Tenu Keimot [a]

[a] Institute of Physics, 142 Riia str., 202400, Tartu, Estonia

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BALLISTIC PROPAGATION OF LUMINESCENCE PULSES IN ANTHRACENE CRYSTAL FLAKES

JAAK AAVIKSOO AND TÕNU REINOT
Institute of Physics, 142 Riia str., 202400 Tartu, Estonia

Abstract Time- and space-resolved luminescence studies of anthracene crystals have revealed efficient radiative energy transfer by polaritons over macroscopic distances at very low velocities ($v<10^{-3}c$). Diffusive and ballistic propagation regimes of (quasi)waveguide polariton modes can be differentiated. Spatial distribution of luminescence intensity reflects the crystal (and defect) symmetry. Anthracene microcrystals may be useful as media for optical signal transfer in molecular structures.

INTRODUCTION

Energy transfer by excitons has been extensively studied by different methods (sensitized luminescence, transient gratings etc.) for several decades. In the present paper we report the results of spatially resolved luminescence studies of energy transfer in a classical molecular exciton system - anthracene crystal. It is characterised by a dipole transition with an oscillator strength $F=0.24$, which brings about strong exciton-photon mixing and, correspondingly, polariton effects. It has served as a model system for excitonic polaritons and as a direct proof of the latter the group velocity experiments[1,2] have demonstrated a drastic slowing down of light near exciton resonances as compared to nonresonant regions. We show further that this slowing down is also observable in luminescence and results in the appearance of ballistically propagating luminescence pulses.

CRYSTAL CHARACTERISTICS AND EXPERIMENTAL SET-UP

The sublimation grown anthracene crystals are thin (.5 to 50 μm) flakes of about 1cm^2 area. The lowest exciton transition is polarized along the **b**-axis in the crystal plane. The dielectric function, which characterizes light (polariton) propagation in the crystal in the luminescence region, contains the following parameters: background dielectric constant ε_o=2.5, oscillator strength F=0.24 and resonance frequency ν_o=25097cm^{-1}. The low temperature luminescence spectrum of the crystal consists of a broad (50cm^{-1}) band immediately below the resonance and its replicas due to optical vibrations. Most of the luminescence is concentrated in the spectral region less the LT-splitting (Δ_{LT}=463cm^{-1}) below ν_o.

The exciting laser pulses of 5ps duration were focused into a 100x500μm spot on the crystal immersed in liquid He at 2K. Luminescence was collected from the rear face of the crystal flake with 20μm spatial resolution and analysed by time-correlated single photon counting technique through a subtractive double monochromator.

PROPAGATION OF LUMINESCENCE PULSES

The kinetics of luminescence at a given wavelength and distance from the excitation spot has a fast response at zero delay and a delayed second maximum, whose intensity and delay ΔT depend on wavelength λ, distance D, and orientation with respect to the crystal **b**-axis (Fig.1).

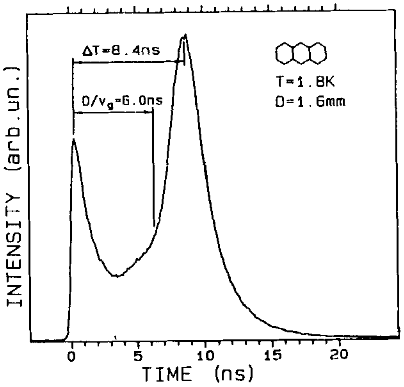

FIGURE 1. Luminescence kinetics of the ν_o-46cm^{-1} line detected at D=1.6mm from the excitation spot.

The experimentally observed linear dependance of ΔT on D and characteristic dependencies on orientation and wavelength suggest that the delayed luminescence maximum corresponds to the luminescence light created at (near) the excitation spot and travelling further ballistically to the detection spot.[3] It is noteworthy, that ΔT as large as 20ns has been observed whereas the luminescence lifetime measured at the excitation spot is τ_o=1-3ns[4,5]. This fact indicates that luminescence light is trapped and survives in the crystal long after the resonant excitonic population has dissapeared. In order to explain quantitatively the observed delay ΔT an initial delay ΔT_o=2.5ns must be added to the delay computed as $\Delta T'=D/v_g$, where v_g is the group velocity at the detection wavelength. This delay is related to the formation, both in time and space, of the ballistic luminescence pulses. The initially created excitations undergo several relaxation (scattering) steps (diffusive propagation) before they end up in a polariton state, the mean free path of which is comparable to the crystal size (ballistic propagation). The value ΔT_o=2.5ns compares well with the excitation spot luminescence lifetime (see above) as well as the (almost exponential) decaytime of the fast component of the luminescence response, therewith supporting the given explanation. At the same time the first, fast luminescence response must be caused by a fast travelling excitation, probably as **c**-polarized component of the exciting light and/or fast surface polariton modes[6], which is transformed into luminescence light at the detection spot.

SPATIAL DISTRIBUTION OF LUMINESCENCE

The spatial distribution of luminescence was studied by analysing the CCD camera image of the luminescing crystal (Fig. 2).

Let us first note the bright excitation spot symmetric with respect to the crystal **b**-axis, strongly emitting crystal edges several millimeters from the excitation spot as well as several brighter spots due to crystal imperfections. This luminescence distribution results from the slab geometry of the crystal and strong exciton-photon mixing. For the strongest luminescence line at ν_o-46cm^{-1} the refractive index n=4.6 and, correspondingly, the angle of total internal reflection ϕ=12.6°. This means that overwhelming portion of the luminescence light, created at the excitation spot, undergo total internal reflection and propagate in (quasi)waveguide modes away from the excitation spot until they are scattered on phonons or crystal imperfections and can escape the crystal.

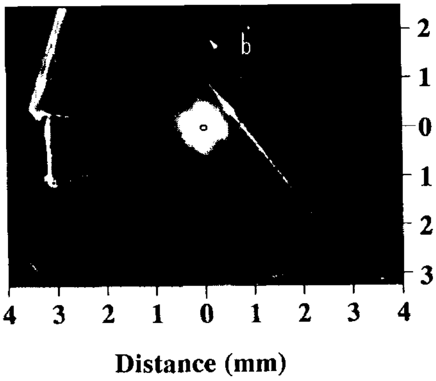

FIGURE 2 CCD camera image of low-temperature (time- and wavelength integrated) luminescence of a thin anthracene flake. The black circle depicts the size of the excitation spot.

Indeed, only about 3% of the total luminescence flux comes from the excitation spot. The cross-section of the integral luminescence distribution is depicted in Fig.3.

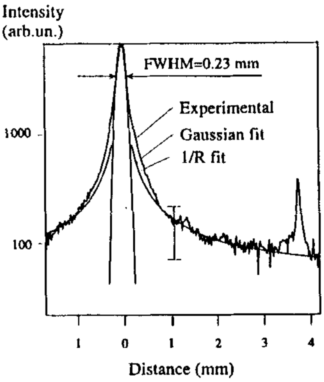

FIGURE 3 The cross-sections of the 2D luminescence distribution along the crystal **a**- and **b**-axis (solid lines). The central part is fitted with a Gaussian and the wings with inverse linear functions (dotted lines). The shape of the excitation spot is given in the middle.

As is evident from Fig.3 two different regions can be differentiated. First, the broadened Gaussian distribution around the excitation spot with a diameter D_D=.3mm and, second, the inversly linear wings, which extend to the crystal edges. We relate these regions to diffusive and ballistic propagation regimes. From the above kinetic results we conclude that the diffusive stage lasts for 2-3ns and further ballistic polaritons propagate at the corresponding group velocities over macroscopic distances until they escape the crystal.

CONCLUSIONS

1. Polariton pulses of a few ns duration propagate ballistically over macroscopic distances in the anthracene flake. Group velocities down to 10^{-3} times the velocity of light in vacuum are observed and the distances involved are tens of mm in the crystal plane. Planar waveguide modes of polaritons are responsible for this propagation.

2. The luminescence trapping phenomenon is observed in the flake crystal. Lifetimes measured from the excitation spot are up to 10 times shorter than the luminescence lifetime of the whole crystal. The total luminescence flux from the excitation spot is about 30 times weaker than the total luminescence output.

3. The propagation of luminescence pulse in the crystal is highly anisotropic. Polariton group velocities along the **b**-axis are 10^2 faster than along the **a**-axis and the anisotropy of the integral luminescence intensity reveals the symmetry of the crystal flake.

4. Gaussian and inverse linear dependence of the luminescence intensity on distance allows one to differentiate between diffusive and ballistic energy transfer.

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