



Temperature-dependent intermolecular coupling and exciton migration in an anthracene containing PPE-PPV copolymer

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ARTICLE INFO

Article history:

Received 2 February 2016

Received in revised form 12 May 2016

Accepted 11 June 2016

Available online 25 June 2016

Keywords:

Photoluminescence

Exciton

Aggregates

Diffusion

ABSTRACT

The dependence upon the temperature of the photophysics of a statistical copolymer anthracene containing poly-(*p*-aryleneethynylene)-alt-poly (*p*-arylene-vinylene)s (AnE-PVstat) as a thin film has been investigated. For this purpose, photoluminescence (PL) spectra have been recorded in a temperature ranging between 10 and 300 K. A modified Franck-Condon model has been invoked to analyze the PL spectra. Our findings point out that upon increasing temperature several vibrational modes are activated starting from a transition temperature leading to molecular rearrangements which leads in turn to a decreased intermolecular distance. The latter fact is consistent with the exciton dynamics which has been found to be governed only by a prompt downhill migration to the exciton band edge at low temperatures up to a transition temperature above which a thermally activated hopping process takes place after the downhill migration.

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1. Introduction

Conjugated polymers are very promising active materials for low cost, ease of production and large area of optoelectronic applications such as organic light emitting diode (OLED) [1,2], organic light-emitting transistors (OLETs) [3], organic field effect transistor (OFET) [4,5] and organic photovoltaic cells (OPVs) [6,7]. Poly (*p*-phenylenevinylene) (PPV) and its derivatives is one of the most promising conjugated polymers due to its high optical and electro optical responses [8,9].

It is well known that an important origin for these optical and electrical properties is the delocalization of π electrons in and between the backbones which leads to the formation of various morphologies such aggregations of polymer chains [10].

Aggregates in polymer films and solution (at low temperature) are intermolecular interactions, which invoke two different classes of interactions. Intrachain interactions lead to J-aggregates; formed when chromophores are oriented in a head to head fashion and interchain interactions lead to H-aggregates; formed

when nearest- neighbor chromophores are oriented in a more side by side manner [11,12].

Photoluminescence and absorption spectra are sensitive to H or J aggregate behavior [13]. For example, in the PL spectrum the ratio of the 0-0 pure electronic intensity to the first vibronic intensity $I_{0-0}^{p-0}/I_{p-1}^{0-1}$, increase (decrease) in H aggregates (J aggregates) with increasing temperature [11–14].

Studies focusing on the temperature dependence of the PL spectra can provide other information than aggregate type such as an exciton diffusion process which is one of the physical phenomena that governs the performance of optoelectronic devices [15,16]. For example, in OPV cells excitons diffuse to the interface between donor acceptor heterojunction where they dissociate into charge carriers. The L_D average distance that an exciton can travel is the diffusion length L_D . This parameter is one of the key parameters determining the internal quantum efficiency of OPV cells [15,17]. The active layer in these devices is often prepared by spin coating or vapor phase deposition technique [9,18]. This facilitates the manufacturing process, but gives rise to disorder effects. A manifestation of this effect is the inhomogeneous broadening σ of the absorption and the PL spectra of thin film. Due to the disorder, excitons diffuse by hopping from one site

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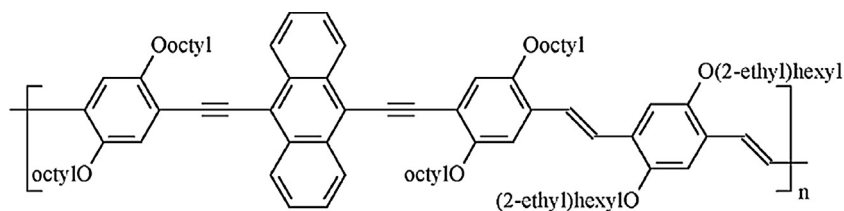


Fig. 1. The chemical structure of the AnE-PVstat.

to the next thereby relaxing energetically and end when they reach the thermal energy level of the most populated states. If σ characterizes the width of the Gaussian distribution of the density of state (DOS) then the thermal equilibrium energy is at $\Delta\varepsilon(T) = -\frac{\sigma^2}{kT}$ [17,19].

Mikhnenko et al. studied, by time-resolved spectroscopy in a conjugated polymer, the temperature dependence in the spectral diffusion and they demonstrated that exciton diffusion is governed by two processes: downhill migration at low temperature, in the range of 4–150 K, and downhill migration with thermally activated hopping at high temperature, in the range of 150–293 K [15]. Furthermore, Hoffmann et al. studied this dependence using comparison between theoretical and experimental dependence of $\Delta\varepsilon(T)$ normalized to $\sigma(T)$ versus $\frac{kT}{\sigma(T)}$ [19].

In this paper, we study the temperature dependence of the PL of AnE-PVstat thin film; whose chemical structure is given in Fig. 1 [20]. The variation with temperature of the ratio 0-0 to 0-1 peak intensity suggests that the AnE-PVstat molecules self-organize to form H-aggregates. A simple Franck Condon (FC) progression is not the adequate modeling, *i.e.* without decoupling the amplitude of 0-0 transition to the rest of FC progression. Hence a modified FC progression with variable 0-0 peak intensity has been used to reproduce the experimental PL spectra. From the fit we have determined the variation of α (related to the amplitude of the 0-0 transition peak), the disorder parameter σ and the electronic transition energy E_0 with temperature.

Through a comparison between the experimental spectral shift $\Delta\varepsilon(T)$ normalized to energetic disorder $\sigma(T)$ and the theoretical $\frac{\Delta\varepsilon(T)}{\sigma(T)} = -\frac{\sigma(T)}{kT}$ versus $\frac{kT}{\sigma(T)}$, we have shown that two exciton diffusion processes are present in AnE-PVstat.

2. Experiments

The material was synthesized and purified as described elsewhere [20]. Thin film of AnE-PVstat was fabricated by spin coating from 10 mg/ml chloroform solution on quartz substrates. The thickness of the spin cast film is estimated to be about 100 nm. The photoluminescence measurements were carried out in a He closed cycle cryostat allowing a temperature variation between 10 K and room temperature. The sample was excited in its absorption band under 488 nm of an Ar⁺ laser with a power of 20 W cm⁻². The spectral response was collected to 250 W cm⁻² Jobin-Yvon monochromator equipped with a photomultiplier. The signal was then diffused on a computer after being treated with synchronous detection.

3. Results and discussion

Fig. 2(a) displays the PL spectra of AnE-PVstat thin film recorded at different temperature. As shown in this figure, for all temperatures, two peaks are observed; the first one originates from 0-1 vibronic transition and the other one related to 0-0 pure electronic transition. It is noted that upon increasing temperature, the PL

spectra blue shift and broaden due to increased thermal disorder. We also remark an increase of the 0-0 peak intensity (Fig. 2(b)).

In order to highlight these observations, the PL spectra were decomposed into two Gaussians emission curves located at 2.165 eV and 1.957 eV, when the temperature is about 160 K, with spacing between each neighbor peak of about 0.197 eV, as shown in Fig. 3(a).

This energy difference is almost similar to the strongest Raman signals around 0.197 eV (1588.3 cm⁻¹) assigned to the stretching mode of the C = C bond.

Like the most of the π conjugated systems, progressions in AnE-PVstat are originated from the coupling with the stretching mode [21,22].

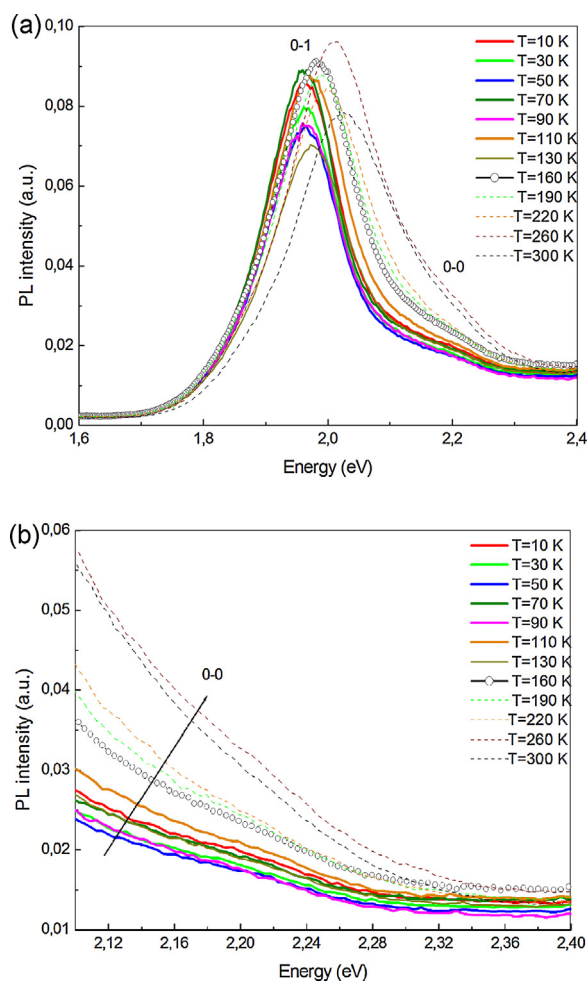


Fig. 2. (a) PL spectra of AnE-PVstat thin film recorded at different temperature and obtained using excitation energy equal to 2.54 eV. (b) The zoom-in spectra highlighting the 0-0 emission peak.

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