Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

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Version of record first published: 04 Oct 2006

To cite this article: Anatoliy Avdeenko, Nina Churakova, Victor Eremenko, Nikolay Gorbenko, Anatoliy Pugachev, Nataliya Silaeva, Yuriy Tiunov & Piotr Zinoviev (1998): Photoluminescence of C_{60} Thin Films at Low Temperatures, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 324:1, 89-94

To link to this article: http://dx.doi.org/10.1080/10587259808047139

Mol. Cryst. Liq. Cryst., Vol. 324, pp. 89–94
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Photoluminescence of C_{60} Thin Films at Low Temperatures

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Photoluminescence spectra of thin films (free and on (001) surface of NaCl wafer) of fullerite C_{60} are studied at 5 K. Film structures were analyzed by standard transmission high energy electron diffraction (THEED) technique. Influence of film structure and substrate on luminescence spectra was observed. It is assumed that the decay of Frenkel excitons on grains boundaries in nanodispersive films competes with energy transport to X-traps and the intensity of excited X-traps photoluminescence strongly decrease. It is supposed that this effect is revealed when a singlet exciton diffusion length is comparable with grains sizes.

Keywords: fullerite C_{60}; low temperature photoluminescence; transmission high energy electron diffraction; X-traps; Frenkel exciton

INTRODUCTION

As a part of intense studies of the physical and chemical properties of the fullerenes (for reviews, see [1] and references therein) photophysical properties of fullerite C_{60} attract unremitting attraction. First of all this interest is bound up with unique symmetry and, as a consequence, with peculiarities of excited states spectrum. Free singlet and triplet excitons are formed[2-7] under optical excitation of fullerite C_{60}. Radiation transitions from the lowest excited states to the ground state are dipole forbidden. Now mechanisms which are responsible for the molecular and crystal luminescence of C_{60} are heavily investigated. So-called X-raps for excitons[10] stipulated by structure

defects and residual impurities play important role in forming luminescence properties of molecular crystals, especially thin solid films prepared by vacuum deposition. A luminescence of excited X-traps in fullerite C_{60} crystals[6, 11-12] considerably complicate own low temperature luminescence spectrum. Recently attempts are undertaken for clearing up of influence of a structure on C_{60} thin film luminescence[6, 13,14]. In this paper we present results on fullerite C_{60} low temperature photoluminescence (PL) using C_{60} thin films. For the first time the PL spectra of the free crystalline thin films were studied. PL spectra of oriented and nonoriented thin films on NaCl substrate were also investigated.

Experimental

Fullerite films were obtained by evaporation of C_{60} single crystals with a purity not worse than 99.9% and condensation onto (100) surface of NaCl wafer having a temperature in the range from 300 to 400 K in vacuum ~ 10^{-3} Pa. The deposition rate was about 0.1 nm s^{-1}. Deposition rate and film thickness were monitored by a quartz oscillator. The films structures was analyzed by standard transmission high energy electron diffraction (THEED) technique. The structure analysis showed that the structure of films depend on a deposition rate, substrate temperature, and film thickness. According to the results of electron diffraction and electron microscopic study, C_{60} films performed on substrate at T ~ 400 K were continuous, ordered, and has an fcc lattice with a period close to that for a bulk sample. Sharp THEED patterns contained reflexes (220) and (422) indicating that the (111) plane of the C_{60} film was parallel to the (100) plane of NaCl wafer. THEED patterns of disordered films performed on substrate at T ~ 300 K have broad halo-type rings. Preliminary analysis shows that these rings can be indicated in fcc structure with following crystallization by the orienting influence of the

substrate. Thus the structural order of deposited C_{60} films can be varied from a highly disordered (or amorphous-like) to a well-ordered (epitaxial growth on a single crystal substrate) state. The free C_{60} films prepared by dissolving substrate in water were located on a copper grid.

The photoluminescence (PL) of C_{60} films were excited by Hg lamp 2.84 and 2.27 eV spectral lines. Measurements were carried out in a liquid helium cryostat at 5 K. The PL spectra were monitored at the right angle to the excitation path with a fast MDR-3 scanning spectrometer covering spectral range from 1.5 to 1.85 eV. Signals were detected by a cooled FEU-79 (S-6) photomultiplier in a standard photon-counting scheme with signal averaging. We corrected PL spectra for the wavelength dependence of the quantum efficiency of the photomultiplier. We perform a deconvolution of the PL spectra by using Gauss line shape for emission bands.

Results and discussion

The C_{60} PL spectra both for the free crystalline thin films and crystalline thin films on NaCL substrate are similar with broad peaks at 1.78, 1.71, 1.6, and 1.52 eV (Fig. 1). This similarity means that NaCl substrate influence on fullerite C_{60} excited states comparatively weakly however intensity of the peak at 1.78 eV decrease for the free thin films. The PL spectra overall profile and peak position don't change depending on the excitation energy. The PL spectra of nonoriented (Fig. 2(a)) and oriented (Fig. 2(b)) C_{60} thin films differ essentially. Corresponding THEED patterns for these films are shown on Fig. 2 (a,b). The grain average size of nonoriented films is about 5 nm. This size corresponds to 3-4 periods of the C_{60} lattice. It means that the films have a extreme dispersibility degree. At such dispersibility more than half of fullerite substance belongs to the boundaries of grains. The grain average size of oriented films is 30-40 nm .

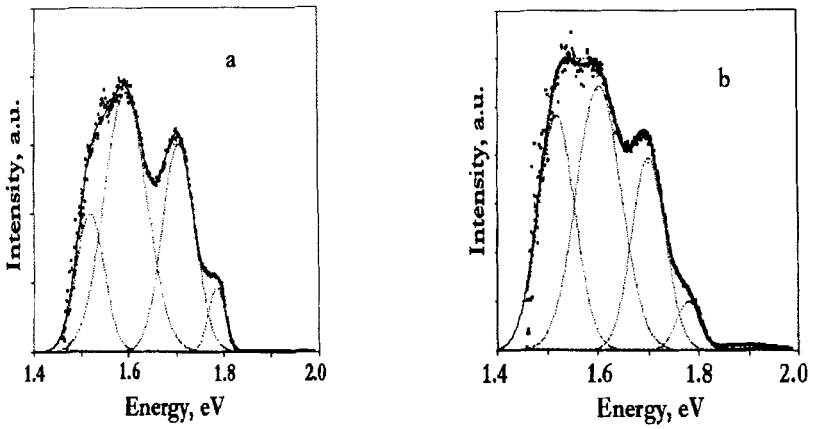

FIGURE 1 PL spectra of oriented C_{60} thin film of 100 nm thick at 5 K after excitation at $h\nu_{ex}$ = 2.27 eV (fits were performed by Gaussians):
a - PL spectrum of C_{60} thin film on NaCl substrate;
b - PL spectrum of free C_{60} thin film.

We have considered the probable versions of nature of excited states of C_{60} crystals. We consider the PL band at 1.71 eV as radiative recombination of a self-trapped exciton of an excimer type without inversion center and the PL band at 1.78 eV as radiative recombination of excited X-traps of fullerite crystal[11, 14]. We assume that in nanodispersive films the radiationless decay of Frenkel excitons on grains boundaries[15] competes with energy transport to X-traps and the intensity of the band at 1.78 eV strongly decrease. It is supposed the effect is revealed when a singlet exciton diffusion length is comparable with grains sizes.

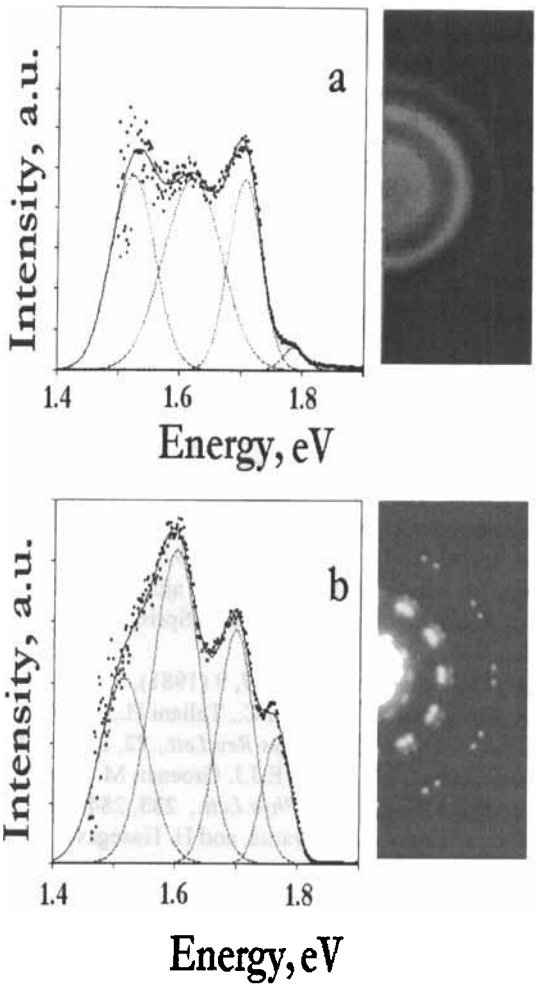

FIGURE 2 Photoluminescence spectra of nonoriented (a) and oriented (b) C_{60} thin films after excitation at $h\nu_{ex}$ = 2.27 eV. Nonoriented thin film of 25 nm thick was prepared with ca. 0.1 nm s^{-1} deposition rate on (001) surface of NaCl substrate at 290 K. Oriented thin film of 20 nm thick was prepared with ca. 0.1 nm s^{-1} deposition rate on (001) surface of NaCl substrate at 373 K. Fits were performed by Gaussians. Corresponding THEED patterns are shown.

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