Materials Letters 168 (2016) 210–213

Contents lists available at ScienceDirect

Materials Letters

journal homepage: www.elsevier.com/locate/matlet

Fabrication and characterization of silver mirror planar microcavity with dye J–aggregates

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ARTICLE INFO

Article history:
Received 13 October 2015
Received in revised form
29 December 2015
Accepted 16 January 2016
Available online 18 January 2016

Keywords:
J–aggregates
Microcavity
Cavity polariton
Vacuum deposition
Organic
Optical materials and properties

ABSTRACT

Silver mirror planar microcavity containing dye J–aggregates is fabricated by a vacuum deposition method. Characterization of the microcavity with the dye J–aggregates of lower concentration (7.70×10^{-4} M) in comparison with the previous studies is performed. Optical strong coupling between excitons and confined photons within the microcavity is demonstrated from the anticrossing of exciton and photon modes. Angle-resolved transmission measurements give a Rabi-splitting energy of ~190 meV for the upper and lower polariton splitting. The fabrication of such a simple metal–mirror–based microcavity (Q–factor ~13) leads to a progress of polaritonic characterization of organic media.

1. Introduction

Optical properties of organic semiconductor microcavity polaritons have become one of a major interest in materials science owing to large oscillator strengths and exciton binding energies [1–4]. The Rabi–splitting energy Ω of ~80–1000 meV [1–3] has been reported for organic semiconductor microcavities at room temperatures. In addition, easy processability of organic materials in air is key to achieve low–cost devices operating at room temperature. Among a variety of organic microcavity materials, J–aggregates have excellent properties such as higher absorbance, narrower spectral widths and smaller Stokes shifts [1,2], as compared with other materials. These characteristics of organic dye J–aggregates are suitable for the cavity polariton formation which is based on repeating process of absorption and emission in the microcavity.

In most of microcavities, the used DBR mirrors with high flatness and strong light confinement are fabricated by sputtering [1] which is not suitable for organic active materials due to unavoidable damages. Meanwhile, there are few reports on fabrication and characterization of cavity mirrors using a vacuum deposition method owing to their insufficient flatness and weak light confinement. In the present study, we have investigated exciton–polariton properties of cyanine dye J–aggregate microcavity using planar silver mirrors fabricated by the vacuum deposition.

2. Experiments

A 50 nm–thick silver layer was vacuum–deposited at a rate of 0.01 nm/s (0–10 nm) and 0.1 nm/s (10–50 nm) onto a quartz substrate. To prepare a dye–aggregate–doped polymer solution, a poly (vinyl sulfate) potassium salt (PVS–K) was dissolved in a distilled water (1.60×10^{-1} M) at 70 °C for 15 min. The PVS–K was mixed with a methanol solution (1.57×10^{-3} M) containing a cyanine dye J–aggregate, 1,1′-Diethyl-2,2′-cyanine iodide (Aldrich), was stirred at 60 °C for 15 min. The dye concentration in the mixed solution is ~7.70×10^{-4} M. Then, J–aggregate–doped polymer films (~400 nm thick) were obtained by spin–coating the solution onto a vacuum-deposited silver layer (50 nm thick) at 500 rpm for 200 s. Finally, optical microcavity was prepared by vacuum–depositing a top silver layer (30 nm thick) onto the polymer/dye film. The slow deposition rates (0.01 nm/s and 0.1 nm/s) allow us to prepare an active layer with less thermal damage during the mirror deposition.

For angle-resolved transmission spectroscopy, a halogen lamp was used as a light source. Transmission spectra were measured by

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http://dx.doi.org/10.1016/j.matlet.2016.01.059
0167-577X/

rotating the microcavity with respect to a charge–coupled device (CCD) spectrometer (Jobin Yvon – SPEX/Horiba Triax 320).

3. Results and discussion

The room temperature photoluminescence (PL) (red line) and absorption (blue line) spectra of the thin film of the cyanine dye J–aggregates dispersed in the PVS–K matrix are shown in Fig. 1. To confirm the formation of the J–aggregates, a dye monomer (isolated molecule dispersed in the solution) solution was prepared by mixing the PVS–K aqueous solution (1.40×10^{-1} M) with the dye solution (4.40×10^{-5} M). The absorption spectrum of the monomer solution is also shown by black line in Fig. 1. In the absorption spectrum of the monomer solution, the absorption band of the monomer appears at ~2.371 eV. In the absorption spectrum of the J–aggregates dispersed in the PVS–K film, a strong and narrow absorption band with a full width at half maximum (FWHM) of ~28 meV appear at ~2.164 eV owing to the formation of the J–aggregates. The absorption peak energy of the monomer was red–shifted due to the intermolecular interaction in the densely doped film, as compared with the monomer solution. The PL band has the FWHM of ~38 meV. The Stokes shift between the absorption and the PL maxima is small (7.1 meV) in comparison with the absorption linewidth.

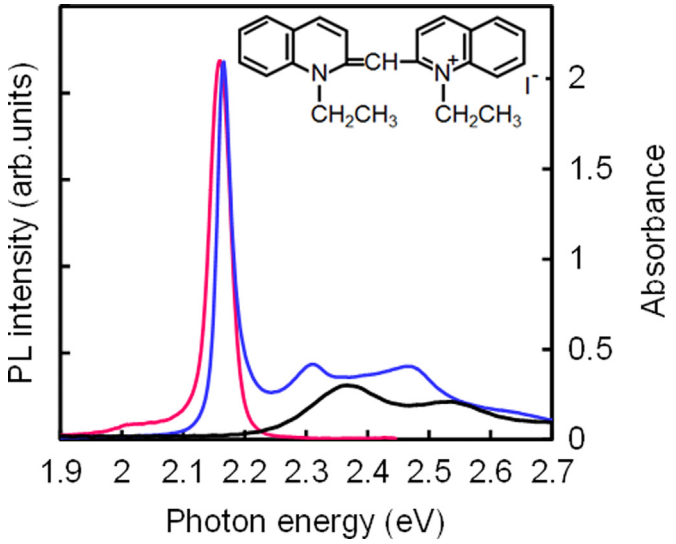

Fig. 1. Optical absorption (blue line) and PL spectra (red line) of the thin film of the cyanine dye J–aggregates dispersed in the PVS–K matrix. A black line shows the absorption spectrum of a monomer solution. The inset shows the molecular structure of 1,1′-Diethyl-2,2′-cyanine iodide. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Fig. 2(a) shows an Atomic Force Microscope (AFM) image of the surface of the thin silver film. Fig. 2(b) shows a transmission spectrum of the J–aggregate–based microcavity composed of a pair of the thin silver films having high flatness (average ± ~7 nm). This surface roughness of ~7 nm is determined by the deposition condition since the surface flatness of the quartz substrate is quite high (~20 Å). Two prominent peaks (red and blue open triangles) appeared at lower and higher energy sides of the exciton energy of ~2.164 eV.

Reflectance (R) of the thin silver film with the thickness of 50 nm was measured using an aluminum flat mirror (R~90%) as a reference to perform the characterization of the fabricated microcavity. As a result, R of ~59% was obtained in the optical absorption energy region of the J–aggregates.

Next, the bare cavity photon mode was determined in order to obtain the upper and lower polariton dispersion curves in the strong coupling regime. We obtained an experimental angle-dependent transmission spectral image indicating photon dispersion curve as shown in Fig. 3. This spectral image was obtained by using the microcavity consisting of an undoped PVS–K film sandwiched between two silver mirrors. The incident light was perpendicular against the cavity, and this angle was set to 0°. The dashed (E_{ex}) and dash–dot (E_{ph}) lines show the exciton energy of the J–aggregates and photon energy of bare cavity photon, respectively. From the absorption spectrum of the PVS–K/dye film in Fig. 1, E_{ex} of ~2.164 eV was used. The cavity photon dispersion curve shown by dash–dot line in Fig. 3 is given by:

$$E_{ph}(\theta) = E_{ph}(0)\sqrt{1 - \sin^2\theta/n_{eff}^2} \quad (1)$$

.where θ is rotation angle and n_{eff} is an effective index [2,5]. We determined the photon dispersion curve (dash–dot line) by fitting to the angle-dependent spectral image by using Eq. (1). In Eq. (1),

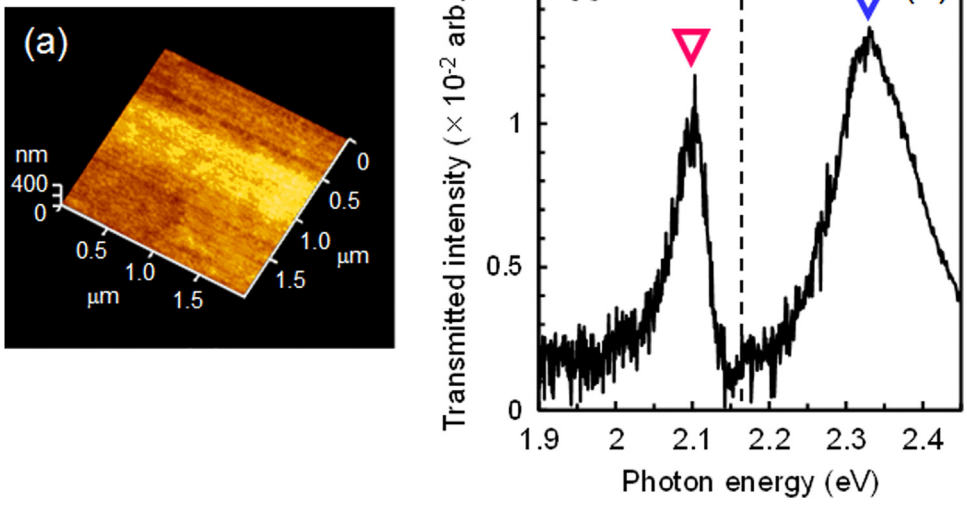

Fig. 2. AFM image of the thin silver film (a) and transmission spectrum of the microcavity containing cyanine dye J–aggregates (b). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)