

Control of Fano line shapes by means of photonic crystal structures in a dye-doped polymer

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(Received 15 November 2006; accepted 23 April 2007; published online 14 May 2007)

The emission of a two-dimensional organic photonic crystal structure is investigated. A photon emitted by a molecule in this structure can take two different coupled pathways. It can either be emitted directly into the surroundings or first be transferred to a discrete state of the photonic crystal structure from where it is then emitted. This unique coupling of the pathways results in a particular asymmetric spectral line shape referred to as Fano resonance. By studying the Fano line shape as a function of the quality factor, the authors can gain insights into the coupling between the pathways within the photonic crystal structure. © 2007 American Institute of Physics.

[DOI: [10.1063/1.2739334](https://doi.org/10.1063/1.2739334)]

There has been considerable interest in the optical and electronic properties of conjugated polymers and molecularly doped polymers owing to their prospective applications in electronic and optoelectronic devices.¹ At the same time, there has been much progress in the rapidly developing field of cavity quantum electrodynamics.²⁻⁴ Within a resonant or off-resonant cavity, fundamental optical processes are modified in a characteristic way, leading to the inhibition or enhancement of spontaneous emission, nearly thresholdless lasing, as well as the modification of the resonant dipole-dipole interaction.^{5,6}

In 1961, Fano proposed that in a system consisting of a discrete state coupled to a degenerate continuum, a transition from or towards a common ground state occurs through two interfering pathways.⁷ One pathway can be described as the direct one from the continuum to the ground state and the other is the indirect one from the continuum through the discrete state to the ground state. The result of this interference is a characteristic asymmetric line shape characterized by the Fano factor f . The Fano factor is the ratio of the transition probabilities of the indirect transition and the direct transition into the ground state. In the limit where one of the two transitions predominates, the Fano factor becomes $f \ll 1$ or $f \gg 1$ and the line shape becomes symmetric.^{8,9} Regardless of the nature of the interactions and transitions involved, the Fano formula and formalism are generally valid, making this effect a universal phenomenon.

This Fano effect has been investigated experimentally in semiconductor superlattices,¹⁰ in nanotubes,¹¹ and in quantum dot structures¹² as well as in waveguides coupled to microcavities¹³ and theoretically in photonic crystal structures¹⁴⁻¹⁶ and in the framework of optical bistability.¹⁶⁻¹⁸

In this letter a photonic crystal (PC) structure with embedded dye molecules is studied. Emitted photons which form a spectral continuum can take two possible pathways. Either the photon is first coupled to the discrete photonic state of the PC and then emitted to the free space or it is emitted directly to the free space and essentially forms a background. An interaction between the continuum and the discrete state gives rise to a superposition of both. Therefore, the system now exhibits a quantum interference and the characteristic spectral line shape. The transition probability of the indirect pathway via the discrete photonic state depends on the quality factor (Q factor) of the PC structure, defined as the resonance energy divided by the resonance linewidth. By probing different areas of the PC structure and because the Q factor of the PC structure varies spatially for these areas, we can now demonstrate that quantum interference is determined by the Q factor.

The PC structures investigated consist of a resist/dye (UVII-HS/Coumarin 6) mixture directly structured by laser interference lithography with a square lattice of air holes, as shown in Fig. 1.

We operate the PC around the Γ point as a two-dimensional second-order Bragg structure with a periodicity of 420 nm. Emission from this structure can take two pathways. Either the photons are indirectly emitted via the photonic state of the PC or directly into free space. The indirect emission is very directional and vertical to the PC plane, whereas the direct emission is emitted into the full solid angle. The second-order Bragg structure is essential because

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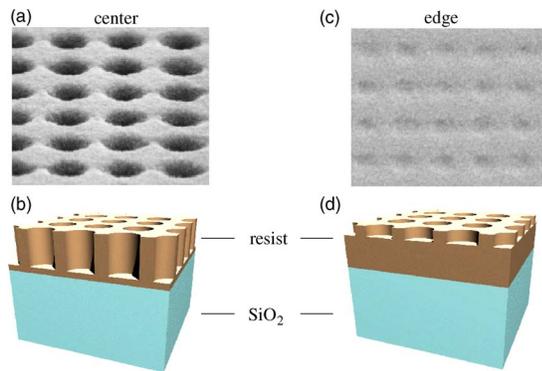


FIG. 1. (Color online) Scanning electron micrograph (SEM) picture of the structure, taken (a) close to the center of the structure and (c) close to the edge of the structure. Schematic view of (b) the geometry close to the center of the structure and (d) the geometry close to the edge of the structure.

both the indirect and the direct pathways have to emit in the same direction. In contrast, for a corresponding first-order Bragg structure, the light that is emitted into the photonic state will remain in the PC structure until it is scattered or absorbed. As a consequence, no interference with the direct pathway can be observed.

The resist/dye mixture was prepared by dissolving 0.8 wt % of Coumarin 6 in the photoresist UVII-HS. This solution was spin coated and post-apply-baked, resulting in an approximately 800 nm thick film. The lattice was generated by two consecutive exposures (wavelength of 257 nm) in a two-beam interferometer separated by a 90° rotation. After exposure, the samples were baked and developed. Because of the Gaussian intensity profile of the writing beams the hole depth varies spatially (see Fig. 1). Therefore, the Q factor of the PC structure varies spatially across the exposed region. The frequency-doubled output of a Ti:sapphire laser at 2.92 eV was used to excite the samples with 80 fs pulses at a repetition rate of 80 MHz. The photoluminescence (PL) emission was collected with a lens system, dispersed with a 600 lines/mm grating, and detected with a nitrogen-cooled charge coupled device camera. To minimize stray light, a 495 nm long-pass filter was used.

Figure 2 shows the comparison between the experimentally measured emission spectrum and the calculated disper-

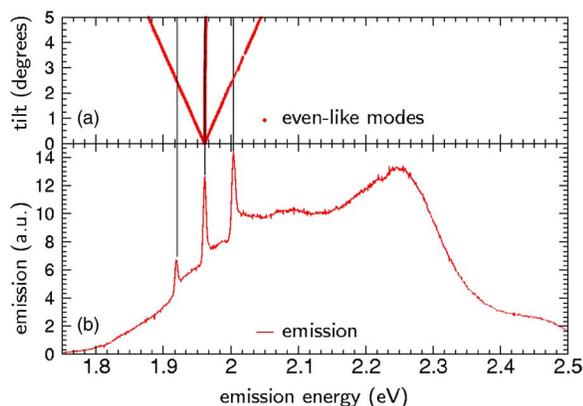


FIG. 2. (Color online) (a) Dispersion relations of the structure. The tilt has been calculated from the k vectors. The Γ point in this graph corresponds to 0°. For a tilt angle of about 2° the measured emission spectrum is perfectly matched. The classification “even-like” corresponds to TE modes; we use this classification because our system is not symmetric with respect to the waveguide plane, i.e., the xy plane. (b) Measured emission spectrum of the structured sample (edge, $T=6$ K).

sion relations of the PC structure. The sharp asymmetric peaks superimposed on the otherwise broad and nearly unstructured PL of the Coumarin/resist mixture provide a clear evidence of discrete photonic states. The dispersion relations of the PC structures have been calculated using the MIT photonic bands (MPB) software package, which performs a preconditioned conjugate-gradient minimization of the block Rayleigh quotient in a plane-wave basis.¹⁹ The calculation cell measures $1 \times 1 \times 11$ periods, and per period 20 plane waves were used. The dispersion relations were calculated for the k vectors that correspond to a tilt of our sample in the cryostat. The threefold degenerate bands split up with increasing tilt angle, e.g., when turning the in-plane k vector from the Γ point towards the X point. Because of the rectangular symmetry of the structure the wave vectors k_x and k_y are indistinguishable and therefore also the rotation around the x axis or the y axis. The coincidence of the sharp resonances observed in the emission spectrum and the ones obtained from the calculated band structure is striking. Furthermore, by comparing the emission spectrum and the band structure, a vertical tilt angle of approximately 2° can be inferred because of the mounting requirements in the cryostat. In the following, we will restrict ourselves to the central resonance mode.

Now we focus on manipulating the ratio of the emitted light between the indirect and direct pathways. The indirect pathway depends on the photonic state. Here, the photonic state is a band-edge state of the PC and its Q factor is a measure for its photonic density. An increasing Q factor indicates an increasing photonic density and therefore an increasing coupling to the photonic state. Hence, the Fano factor f becomes a function of the Q factor, $f \rightarrow f(Q)$. Because of the Gaussian shape of the interference lithography setup, the Q factor of our structure varies spatially. By scanning our excitation laser across the PC structure, we can probe the change in the ratio between the two pathways and therefore the coupling between the two pathways by monitoring the Fano factor f of the asymmetric line shape. The Fano factor f is determined with dependence on the Q factor by fitting the measured spectral line shape for different spatial positions to the Fano profile,

$$F(\epsilon) = \frac{(\epsilon + f)^2}{\epsilon^2 + 1}. \quad (1)$$

Here, $\epsilon = (E - E_R) / (\Gamma_{\text{res}}/2)$, E is the emission energy, E_R is the resonance energy, and Γ_{res} is the resonance linewidth with $Q = E_R / \Gamma_{\text{res}}$. To avoid distortion of the results, here the asymmetry of the peaks, prior to fitting the data, the background taken from the emission spectrum of the unstructured part of the sample is divided. Furthermore, in what follows the Q factor was taken from the fit. Owing to the noise in the measured data, the fit exhibits an uncertainty of about 10% for both the Fano and the Q factors. In Fig. 3(a) the measured line shape at the edge of PC structure and its Fano fit are shown.

For the edge the fit results in a Fano factor f of 3.0 and a Q factor of around 430. Next the Q factor is decreased by measuring the line shape at the center of the PC structure. For the center the fit results in a Fano factor f of 1.40 and Q factor of approximately 300. The asymmetry of the line shape is much more pronounced. As a result of the increasing hole depth and the associated thinner nonstructured polymer/AIP license or copyright, see <http://apl.aip.org/apl/copyright.jsp>

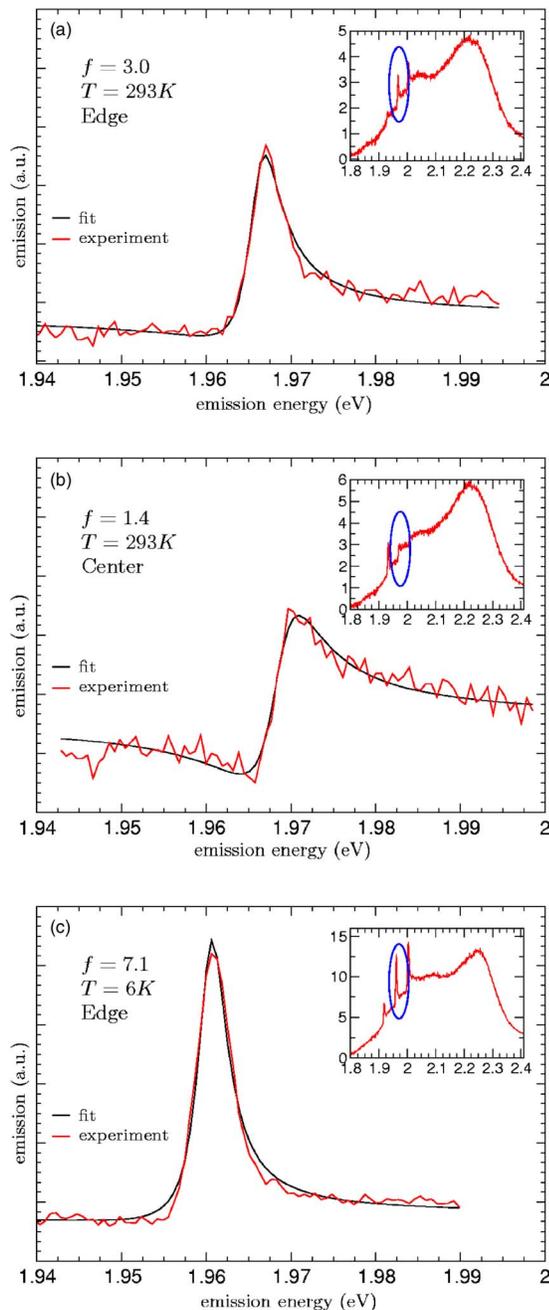


FIG. 3. (Color online) Experimental data and fit of the Fano peak for three different experiments. The emission spectrum is displayed in the insets. (a) Edge of the PC structure, $T=293$ K. (b) Center of the PC structure, $T=293$ K. (c) Edge of the PC structure, $T=6$ K.

dye layer underneath the PC structure, the light is guided more weakly and therefore the Q factor decreases towards the center of the PC structure. Furthermore, this gives rise to a slight energy shift of the resonances because the effective refractive index of the guided mode also varies with the depth of the holes. The experimental data clearly prove that in such PC structures the Fano parameter and therefore the strength of the quantum interference can be tuned by the Q factor.

Another way to tune the Q factor in our system, and therefore the Fano factor, is to vary the temperature. A temperature change leads to a change of the refractive index, which again leads to a change of the Q factor. For the edge of the PC structure the measured line shape and its Fano fit are shown in Fig. 3(c) for a lower temperature of 6 K. The

comparison with a temperature of 293 K [Fig. 3(a)] reveals that the Q factor stays almost constant within the range of inaccuracy (increases slightly from 430 to 460), the Fano factor f increases from 3.0 to 7.1, and the peak shifts by 0.007 eV to a slightly lower energy. A higher Q factor and a lower peak energy corroborate the increase of the refractive index. Taking into account that the peak shifts by approximately 0.4%, that about a third of light is located in the polymer/dye layer, and that the change of the refractive index of the underlying SiO_2 substrate is negligible, the refractive index of the polymer/dye layer increases to approximately 1.2% when the sample is cooled. This agrees very well with the measured thermo-optic coefficients dn/dT of around $-1 \times 10^{-4} \text{ K}^{-1}$ (Ref. 20) for typical polymers.

To exclude contributions due to stimulated emission, we cross-checked our experimental results by varying the excitation intensity. All data presented are well below the threshold for stimulated emission, which therefore can be safely ruled out as a contribution leading to the observed quantum interference.

In conclusion, Fano emission line shapes are observed from a PC structure with a dye-doped resist as active material. While the Fano coupling is a universal phenomenon in physics and has been observed in a variety of physical systems, here a tunable organic Fano system is realized. Controlling the Fano line shape by manipulating the pathways responsible for quantum interference has been demonstrated either by varying the Q factor of the PC structure or by changing the temperature T . Further development of this field of quantum electrodynamics will undoubtedly open a route towards more complex structures for both further fundamental investigations as well as practical applications such as optical switches and sensors.

The authors gratefully acknowledge W. Bächtold, T. Stöferle, and S. Gulde for stimulating discussions, and the Swiss Federal Innovation Promotion Agency (KTI) for financial support.

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