Experimental demonstration of Fano-type resonance in photoluminescence of ZnS:Mn/ SiO2 one-dimensional photonic crystals

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Experimental demonstration of Fano-type resonance in photoluminescence of ZnS:Mn/SiO₂ one-dimensional photonic crystals

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We investigated the photoluminescence (PL) properties of ZnS:Mn/SiO₂ one-dimensional photonic-crystal structures. PL spectra were measured from two opposite directions perpendicular to the sample. The cavity mode emission measured from the sample surface showed asymmetric spectral shape; measurements from the back side of the sample showed a symmetric spectral shape. The experimental spectra were analyzed by a simple model calculation based on the transfer matrix method. From the model calculation, it was found that the asymmetric shape observed in cavity mode emission is caused by Fano-type resonance which is the coupling effect between discrete emission from the ZnS:Mn cavity layer and continuous background emission from all ZnS:Mn layers except the cavity layer. © 2005 American Institute of Physics. [DOI: 10.1063/1.2117611]

Since the first demonstration of epitaxial semiconductor Bragg reflectors for surface-emitting laser diodes, which was followed by the first realization of distributed feedback surface-emitting laser diodes with multilayered heterostructures, photonic crystals (PCs) have drawn much attention as a new kind of optical material. The PCs—artificial low-loss periodic dielectric materials—are characterized by photonic band gaps (PBGs). The well-known effect of PBGs is that if the wavelength of the incident light is within the PBGs, the light is totally reflected by the PCs. Also, PBGs have the ability to alter the spontaneous emission from radiation sources by means of electromagnetic mode distributions, different from that of the free-space mode distribution. So, interesting phenomena, such as photon-atom bound dressed states, nonexponential decay of spontaneous emission, strong inhibition of emission, and enormous enhancement of radiation, have been theoretically and experimentally reported by several groups. And recently, nonlinear effects using PCs, such as second-harmonic generation and the optical bistability effect, are being intensely investigated by some groups since these are promising for optical circuits. Nonlinear optical bistability in a zero-dimensional cavity and one-dimensional waveguide system, which causes the Fano resonance effect, was theoretically proposed by Cowan et al. But, as far as we know, such Fano resonance effects have not ever been observed experimentally because of the difficulty in fabricating a multidimensional PC structure. In this letter, we report on the Fano-type resonance in the luminescence spectra of a ZnS:Mn/SiO₂ one-dimensional PC. This structure, the background emission from all of the ZnS:Mn layers, except the cavity layer, forms a broad spectral mode corresponding to a continuous state, while the emission from the cavity layer forms a sharp mode corresponding to a discrete state. If the cavity mode couples with the background mode, we can observe the Fano-type resonance which gives rise to an asymmetric spectral shape in the photoluminescence (PL) spectra.

We fabricated a microcavity structure which consists of alternating ZnS:Mn (Mn:0.5 wt %) and SiO₂ layers, as shown in Fig. 1. These two materials were deposited on a glass substrate by a radio-frequency-sputtering method at room temperature. The refractive index of the ZnS:Mn and SiO₂ are 2.5 and 1.5, respectively. The thickness of the layers was controlled by deposition time. The thickness of the ZnS:Mn layers are 58 nm, and that of the SiO₂ layers are 97 nm. There are 21 layers in total, and the ZnS:Mn cavity layer with the thickness of 116 nm is centered in the multilayered structure.

Room-temperature PL measurements were performed perpendicular to the sample at room temperature. The sample was excited by the 325 nm line of a He–Cd laser. The laser light was focused on the sample surface, and PL light was detected from two opposite directions, that is, from the sample surface and from the back side of the sample as shown in Fig. 1. The former is like a reflection measurement—“Arrangement 1,” and the latter is like a transmission measurement—“Arrangement 2”. Reflectance spectra were measured using a conventional spectrometer with an incident angle of 5°.

In Fig. 2, we show the normalized PL spectra measured in Arrangements 1 and 2. In general, ZnS:Mn shows a broad emission band at around 590 nm, which originates from the d-d transition of Mn²⁺ ions. Such a broad emission band can be modified by the multiplayer microcavity structure. In

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FIG. 1. A schematic picture of sample structure and PL measurement.
order to clearly see the effects of the microcavity structure, we normalized the raw PL spectra, \(I_{PL}\), by the measured PL spectrum of a single thick ZnS:Mn layer (about 1.5 \(\mu m\)), \(I_{Mn}\), as \(I_{norm}=I_{PL}/I_{Mn}\). As shown in Fig. 2, the normalized PL spectra show a sharp emission at 580 nm, which corresponds to the cavity mode, and the position of the cavity mode emission is consistent with the reflection spectrum. In addition, one can see broad structures in the longer-wavelength region between 600 and 700 nm. From a comparison between Arrangements 1 and 2, we can see clear differences in the features of the two normalized PL spectra. Although the PL spectrum is like a mirror image of the reflection spectrum in the case of Arrangement 2, the normalized PL spectrum in the case of Arrangement 1 shows some peculiar features. The emission is observed even in the stop band between 590 and 650 nm, and the broad emission peaks—not at the dip position—but in the middle of the sharp reflection edge in the reflection spectrum. The most remarkable feature of the difference between the spectra of Arrangements 1 and 2 is the asymmetric shape of the cavity mode emission at 580 nm. The spectral shape of the cavity mode emission is symmetric in the case of Arrangement 2; however, it is asymmetric in the case of Arrangement 1. In this study, we shall concentrate on the asymmetric shape observed in the cavity mode emission of Arrangement 1.

In order to understand these observations, we constructed a simple model to analyze the emission spectra. In our model, light sources originate from every ZnS:Mn layer, and the emission from each layer is considered to be plane waves. The most important points in this model are the consideration of multiple reflections at the interfaces and the exponential decay of excitation laser intensity that causes differences between Arrangements 1 and 2. First, we consider the emission from one of the internal ZnS:Mn layers. From the light source, the plane wave starts to propagate toward the upper and lower layers. After multiple reflections at the interfaces, the light will exit the sample structure. The electric fields of the plane wave which start toward the upper layers and lower layers, A and B, respectively, can be expressed as follows:

\[
\sum_{i=1}^{\infty} A(i) = t_1 \left[ 1 + r_1 r_2 \exp \left( -\frac{i4\pi n_1 d}{\lambda} \right) \right]
+ \left\{ r_1 r_2 \exp \left( -\frac{i4\pi n_1 d}{\lambda} \right) \right\}^2
+ \left\{ r_1 r_2 \exp \left( -\frac{i4\pi n_1 d}{\lambda} \right) \right\}^3 + \cdots
\]

\[
\sum_{i=1}^{\infty} B(i) = t_1 t_2 \left[ 1 + r_1 r_2 \exp \left( -\frac{i4\pi n_1 d}{\lambda} \right) \right]
+ \left\{ r_1 r_2 \exp \left( -\frac{i4\pi n_1 d}{\lambda} \right) \right\}^2
+ \left\{ r_1 r_2 \exp \left( -\frac{i4\pi n_1 d}{\lambda} \right) \right\}^3 + \cdots
\]

where \(r\) and \(t\) are reflectance and transmittance (for electric field) of multilayers sandwiching the emission layer being considered. These are determined by the transfer matrix method\(^{11}\) for the upper layers \((r_1, t_1)\) and for the lower layers \((r_2, t_2)\). \(\lambda\) is the wavelength of the emitted light, \(d\) is the thickness of ZnS:Mn single layer, and \(n_1\) is the refractive index of the ZnS:Mn layer. Then, the emission from one of the internal ZnS:Mn layer is expressed as

\[
E(k) = [\sum_{i=1}^{\infty} A(i) + \sum_{i=1}^{\infty} B(i)] \exp(-\alpha m d_{ZnS:Mn}).
\]
continuous emission over the wavelength range of the stop band; however, the broadband emission of Arrangement 2 is completely suppressed in the stop band range. In general, when there is a coupling between discrete and continuous states, Fano-type resonance occurs, and the spectrum takes on an asymmetric shape. Accordingly, the asymmetric spectral shape of the cavity mode in Arrangement 1 can be understood as the result of Fano-type resonance between the discrete emission from the cavity layer and the continuous background emission from all the ZnS:Mn layers, except the cavity layer. This situation is similar to the Fano resonance between transmitted light through a zero-dimensional cavity and a one-dimensional waveguide, which was proposed by Cowan et al.\textsuperscript{9} In the case of Arrangement 2, the continuous background emission from all of the ZnS:Mn layers, except the cavity layer, is suppressed in the stop band range. We found that this is because emissions from the upper layers are dominant due to the absorption of excitation laser light from layer to layer. The lack of a continuous emission band in Arrangement 2, which leads to an inhibition of the Fano-type resonance, results in the symmetric spectral shape of the cavity mode emission.

To observe Fano-type resonance, the emitted light needs coherence, and the Fano-type effect causes a phase shift between the discrete and continuous states. In Fig. 4, we calculated the emission spectra considering interference effects of emissions from each layer and the phase shift between the emission light from the cavity layer and the emission from the background layers in order to include the Fano-type resonance. One can see that the asymmetric (Arrangement 1) and symmetric (Arrangement 2) features of the cavity mode emission are well reproduced by this model. The Fano-type resonance, which is represented by the asymmetric spectral shape, arises in Arrangement 1 (solid line). But in Arrangement 2 (dotted line), that resonance does not occur. However, considering that the light is emitted spontaneously from all Mn\textsuperscript{2+} ions, it is expected that the emissions cannot interfere with each other, that is, the emission light is incoherent. However, the PL spectra—which show Fano-type resonance between the cavity mode emission and the other layers’ background emission—suggest the coherent nature of the emission light. At presence, we do not have clear evidence about the origin of this. However, considering that each Mn ion emission is affected by every other mode that exists in this finite size structure, it is probable that these two emissions (from cavity and other layers) have coherence with each other.

In conclusion, we observed Fano-type resonance in the PL spectra of ZnS:Mn/SiO\textsubscript{2} one-dimensional PCs. The different spectral shapes of the cavity mode emission between Arrangements 1 and 2 have been qualitatively explained by simple model calculations. From the experimental results and the model calculations, we found that the emission from the cavity layer has coherence with other background layers’ emission.

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FIG. 3. Calculation results of emission from ZnS:Mn layers, except cavity layer (dotted line), and emission from only cavity layer (solid line) in Arrangement 1 (a) and Arrangement 2 (b). In Arrangement 2, background emission (dotted line) is suppressed.

FIG. 4. Calculated emission spectra of the ZnS:Mn/SiO\textsubscript{2} one-dimensional PC using the Fano resonance formula. In this case, we consider the interference effect between discrete cavity mode and continuous background mode.