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Enhancement of excited-state emission of InAs/GaAs quantum dots with large-period photonic crystal

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Abstract: In this study, the photoluminescence spectra of InAs/GaAs quantum dots material with photonic crystals were investigated by laser diode excitation. The photonic crystals were fabricated in the material of InAs/GaAs quantum dots by laser holography and wet etching method. It was found that the spectra from quantum dots with photonic crystals appeared multi-peak structure; the enhancement and modification to the short-wavelength component were more pronounced than those to the long-wavelength components. The photoluminescence from InAs/GaAs quantum dots was modified by photonic crystals, and the emission from excited states was significantly enhanced.

Key words: quantum dots, photonic crystal, laser holography, photoluminescence spectra
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利用大周期光子晶体结构增强InAs/GaAs量子点的激发态发光

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摘要:在该研究中,通过激光全息和湿法腐蚀的方法在InAs/GaAs量子点材料上制备光子晶体,研究了由激光二极管激发制备了光子晶体的InAs / GaAs量子点材料的光致发光光谱.发现具有光子晶体的量子点材料的光谱显示出多峰结构,光子晶体对短波长部分的发光增强和调制比对长波长部分的增强和调制更明显.InAs / GaAs量子点的光致发光光谱通过刻蚀形成的光子晶体结构得到了调控,并且量子点的激发态发光得到了明显增强.

关 键 词:量子点;光子晶体;激光全息曝光;光致发光光谱
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Introduction

Semiconductor quantum dots (QDs) is a major semiconductor material for both device applications and fundamental physical studies of low-dimensional structure material[1]. Due to the three-dimensional confinement ef-

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fect of charge carriers, the performance of QDs lasers is better than that of quantum well lasers. QDs lasers have narrower gain line, larger peak gain and smaller threshold current density[2]. As reported before for InGaAs QDs, several electron and hole energy levels would be occupied under high excitation. In the spectra, multi-peak features might appear due to exciton recombination from several energy levels[3]. The exciton would be excited for different center wavelength. By investigating the relationship between QDs structure and confined exciton energy levels, the origin of transitions in ground state and excited state could be well understood[4]. It was reported that the degeneracy factor in the excited state is higher than that in the ground state[5]. Therefore, high photoluminescence (PL) efficiency and high output power laser have been obtained from transition in excited state[6-7]. The extraction efficiency of QDs can be enhanced by photonic crystals (PhCs) and micro-structure. A large enhancement by PhCs to PL from quantum dots was attributed to a combination of high-intensity fields and strong coherent scattering effects because of leaky eigen modes of the PhCs. These effect could be achieved by engineering leaky modes and overlapping them with the absorption and the emission wavelengths of the QDs[8].

In this study, we investigated the modification of QDs' spectra by PhCs. We fabricated two-dimensional PhCs on the film of InAs/GaAs QDs by holography[9] and wet etching. The Photoluminescence (PL) spectrum of InAs/GaAs QDs with PhCs were measured and analyzed. It was found that the PhCs we made could modify the spectral structure, and the enhancement factor to the excited-state emission was larger than that of the ground state of the QDs. This asymmetry enhancement was analyzed by comparing the characterization of the excited state and the ground state in QDs, combining with the PhCs bandstructure. The multi-peaks in the spectra resulted from different order excited-state emission. In our experiment, different spectral peaks were attributed to the ground-state emission (longest spectral peak) and different order excited-state emission, respectively. On the PhCs, the excited-state emission was enhanced. The enhancement factor for the excited-state emission was larger than that for the ground-state emission. The reason can be explained in two aspects: one is to explain by the bandstructure of the PhCs we used. Around the wavelength of the pump light and the wavelength of the excited-state emission, there are some leaky modes, which enhance the excited-state emission. Moreover, as the pump light interacts with the QDs, the radiative rate will be enhanced due to Purcell effect of PhCs. Larger is the radiative rate, stronger is the PL intensity. Another may be due to that the degeneracy of the excited state. Usually, the degeneracy of the excited state is higher than that of the ground state. Due to the high PL efficiency related to high degeneracy of the excited state, the enhancement for excited state is larger than that for ground state.

1 Experiment

1.1 Fabrication of PhCs in the InAs/GaAs QDs material

In the experiment, a 10 nm active layer of InAs/GaAs QDs was epitaxial grown on the N-type GaAs substrates by molecular beam epitaxy (MBE). The structure of the GaAs/InAs material is shown in Figure 1(a). The InAs/GaAs QDs layer was embedded between two intrinsic GaAs layers. There are five layers of QDs in the material. A PhCs pattern was fabricated to the material by holography and wet etching. Firstly, a layer of photoresist was coated on the GaAs surface by spin-coating method. After spin photoresist on the GaAs, the material was baked on the hotplate at 100℃ for five minutes. Secondly, PhCs were defined on the photoresist by using photoholography method with He-Cd laser at wavelength 325 nm. After development, PhCs pattern formed on the photoresist on the GaAs surface. Thirdly, the GaAs material was wet etched with the photoresist mask by using phosphoric acid and hydrogen peroxide as etchant solution. The percentage of the phosphoric acid and that of the hydrogen peroxide were both 2.5% in this mixed solution. The etching rate of GaAs/InAs material was 1.46nm/s by using the acid etchant solution. It took approximate 90s to etch awaythe p-GaAs, i-GaAs layer and the InAs QD layer through the photoresist. At last, after removing photoresist, the PhCs with QDs active layer were obtained. A picture of fabricted PhCs is shown in Figure 1 (b), where the period is 3.75 μm, the radius of the hole is approximately 0.68 μm, and the hole depth is around 130 nm.

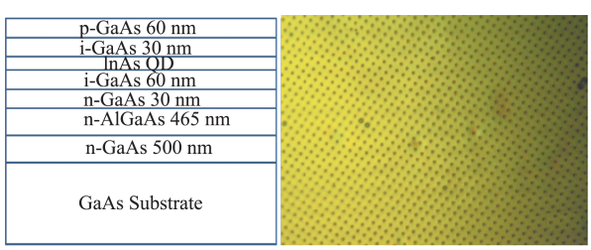

Fig. 1 (a) The structure of InAs/GaAs QDs material. The QDs layer is consisted of InAs quantum dots. The QDs layer is 90 nm under the surface. (b) Picture of fabricated PhCs of GaAs with InAs/GaAs QDs. The period of PhCs is 3.75 μm

图1 (a)InAs/GaAs量子点材料结构.量子点层距离顶层90nm.(b)带有InAs/GaAs量子点的GaAs材料上制备的光子晶体照片.光子晶体周期3.75μm

1.2 The spectra pumped with 10 × objective

The InAs/GaAs QDs was excited by a 400 nm semiconductor laser, the excitation light was focused by a 10×/0.25 objective onto the sample and the PL was collected by the same objective to an InGaAs photodiode detector through a monochromator. Spectra with different pump distances between the sample and the objective in three different areas are shown in Figure 2. The spectra

of the area without PhCs, the spectra of the etched square and the spectra with PhCs are shown in Figure 2 (a), (b) and (c), respectively. The measured spectra could be fitted into multi-Gaussian function:

$$y = y_0 + \sum_i \frac{A_i}{w_i \sqrt{\pi/2}} \exp\left(-2 \times \left(\frac{x - x_{ci}}{w_i}\right)^2\right)$$

where x_{ci} *re*presents central wavelength, w_i represents spectral width, and A_i represents the amplitude.

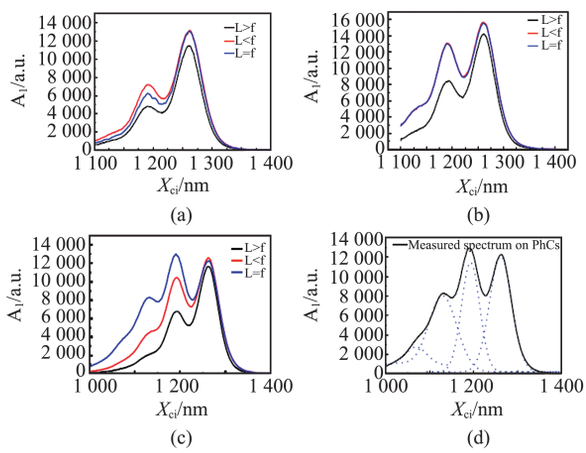

Fig. 2 Comparison of the spectra of QDs in different areas with different objective work distance with 10×/0.25 objective with pump power 19.4 mW. (a) Spectra in the area without PhCs. (b) Spectra in the area of an etched square (etching time same to the PhCs). (c) Spectra in the area with PhCs. The black line represents the spectrum with longest distance between the sample and the objective. The blue line is the spectrum in focus state. The red line is on behalf of the spectrum with middle distance, which is between the longest distance and that of the focus state. The symbol of *L* represents pumping distance, and *f* represents focal distance. (d) The spectrum in the area with photonic crystal in focus state, the blue dotted-lines are fitted lines by multi-Gaussian function.

图2 对比在不同物镜工作距离情况下量子点发光光谱的变化.(a)没有光子晶体区域的光谱.(b)方形的腐蚀区域(腐蚀时间与光子晶体一致).(c)光子晶体区域.黑色的曲线代表样品和物镜距离最远情况下的光谱测试结果.蓝色曲线代表物镜在聚焦状态下的光致光谱测试结果.红色曲线代表的测试光谱曲线的位置代表物镜距离样品的位置在最远和聚焦状态中间.图例中的*L*代表物镜镜头距离材料表面的距离,*f*代表聚焦距离.(d)在光子晶体区域聚焦状态下的多高斯函数拟合的光谱

In Figure 2(a), spectra have only two main spectral peaks even in focus state. The spectra in Figure 2(b) were measured from an etched square, there are two main spectral peaks and a weak peak in the short-wavelength region. The PL intensity of the spectral peak around 1 191 nm at three different distances in Figure 2(b) is enhanced compared to the corresponding spectra in Figure 2(a). In Figure 2(c), there are four spectral peaks in the spectra in the PhCs area. The spectrum in the area of PhCs in focus state was fitted into multi-Gaussian function. In the no-structure area (no etched square and no PhCs), the distance between sample and objective was changed to measure the spectra in the same position. From the measurement results, all the detected spectral lines in the same position with different pump distances are similar, and the spectra in the short-wavelength region are flat. In the etched areas, the PL intensity of the short-wavelength component was improved compared with that in the no-structure area. For comparison of the spectra in focus state with different pump distances, the spectral parameters by fitting the spectra into multi-Gaussian function are listed as following. For the spectrum in focus state of no-structure area without PhCs, the short peak wavelength is 1 192 nm, and the spectral width is 57.9 nm; while the long peak wavelength is 1 262 nm, and the spectral width is 41.7 nm. For the spectrum of the etched square area, a short wavelength peak is 1 192 nm, and the spectral with is 42.0 nm, while the long peak is at wavelength 1 260.8 nm, and the spectral width is 48.8 nm. For the spectrum with PhCs in Figure 2(d), two spectral peaks at long wavelength are 1 193.2 nm, 1 261.5 nm, and corresponding spectral widths are 41.4 nm and 50.6 nm, respectively. The spectral peak wavelength and the spectral width change much as the excitation position changed for long wavelength component. This situation changed for the shorter wavelength. There are only two wavelength peaks in the spectra for the blank InAs QDs material without PhCs. For the spectrum in square area in Figure 2(b), it can be found that there is a shorter broad wavelength peak at 1 133 nm with spectral width of 59.6 nm. The spectrum of the area with PhCs in focus state could be fitted into four-Gaussian function, two short wavelength peaks are 1 069 nm and 1 136 nm, and the corresponding spectral widths are 49.3 nm and 61.4 nm. In the PhCs area, the longest-wavelength component at peak wavelength 1 261 nm has little change compared with those in Figure 2(a) and (b). However, the PL intensities of the middle-wavelength component at peak wavelength 1 193 nm and the shortest-wavelength component have been improved obviously. Usually, the PL intensity of the spectrum in the focal state is strongest. One reason is the pump intensity changes with the pump distance. Another reason is that the collection angle of the PL changes with the pump distance. It means that the spectra of QDs with PhCs are related to the dispersion of PhCs slab. In the experiment, the spectra of no-etched area with different pump power were measured. Similar to those in Figure 2(a), there were only two spectral peaks in the spectra under various pump powers we used. It was found that the spectral peak wavelength and the relative spectral intensities of the short-wavelength region didn't change with pump power changing (below 19.4 mW). It meant that the pump power we used would not affect the numbers of spectral peaks and the intensity of the short wavelength region. The intensity of the short wavelength region of

the spectra in Figure 2(c) would be improved by PhCs .

The PL intensities of the two short spectral peaks of QDs with PhCs were higher than those in blank material and etched-square area at corresponding wavelength. As shown in Figure 3(a), the enhancement factor as function of wavelength was obtained from the ratio of the spectral intensity of the QDs with PhCs in focus state to that without PhCs, indicating that the enhancement factor at short wavelength is larger than that at long wavelength. The enhancement factor is less than 1 around 1 260 nm. It increases slowly as the wavelength decreases from 1 240 nm to 1 100 nm, and it reaches to approximately 7 at around 1 100 nm. It indicates that the QDs' spectrum is enhanced to approximately 7 folds in short wavelength region around 1 100 nm by the PhCs, while the spectrum in long wavelength of region around 1 260 nm have almost no enhancement by PhCs. The longest wavelength peak and the second long wavelength peak, both of them appearing in all the three cases in the spectra in Figure 2, result from the ground-state emission and the first excited state emission, respectively. The third peak around 1 193 nm in Figure 2(b) and (c), and the forth peak around 1 069 nm in Figure 2(c) come from the second excited state emission and the third excited state emission, respectively. It indicates that PhCs enhanced the excited state emission and high-order excited state emission of InAs/GaAs QDs.

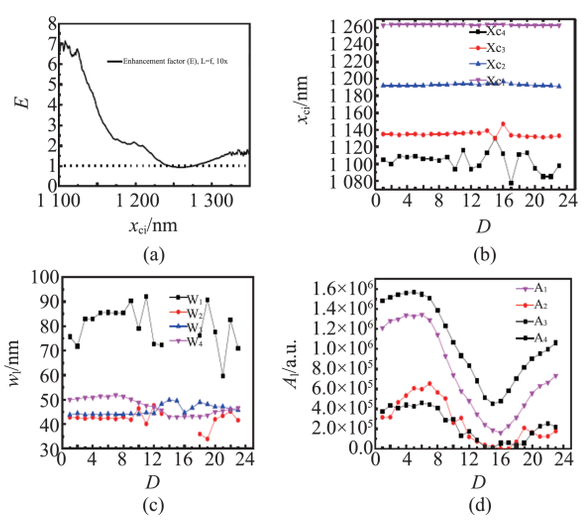

Fig. 3 The spectral parameters from the spectra of QDs in PhC area in different pump distance (D) with 10×/0.25 objective. (a) Enhancement factor (E) as function of wavelength, (b) central wavelength, (c) spectral width, and (d) spectral amplitudes, at different relative pump distance. The position 6 is near the focus position, and the small number means the distance between the sample and the objective is small

图3 光子晶体区域使用10×/0.25物镜在不同距离测试得到的光谱参数:(a)增强因子,(b)中心波长,(c)光谱宽度,和(d)光谱幅值,均为在不同泵浦距离下的测试结果.图中横轴数字6代表样品接近焦点位置,数字小代表样品距离物镜近

The measured spectra of QDs in PhCs area with different pump distances were fitted into multi-Gaussian function, and the obtained parameters of the spectral central wavelength, the spectral width and the amplitude are shown in Figure 3(b)~(d). When the pump distance varies from far to near (from point 23 to point 0), the central wavelengths of the three longer-wavelength components of the spectra almost maintain as constant, while the central wavelength of the shortest-wavelength component of the spectra has some scattering points, which may be caused by background noise because of the low spectral intensities of the shortest-wavelength component. The spectral widths of the two long-wavelength components are around 40~50 nm at different pump distances. The spectral width of the second peak around 1 140 nm is also around 40 nm, and the spectral width of the shortest-wavelength component is around 80 nm, showing large fluctuation. Moreover, the spectral width of two short wavelength components shows singularity at some pump distances (such as points 7, 8, 9, 10) and they are not shown in the figure, which is due to that the spectral intensities are weak at these pump distance as shown in Figure3(d). The spectral intensities for different spectral components at different pump distances show a similar varying tendency. As the pump distance changes from far to near (point 23 to 0), the PL intensity first decreases (from point 23 to 15), then increases (from point 15 to 6), and finally decreases again (from point 6 to 0). Why is the PL intensity still increasing when the pump distance becomes far away from point 16 to 23, the detail reason is not so clear. This might be due to the effect of the QDs slab structure, and the output angle of the slab and the collection angle of the objective, which are affected by the presence or absence of a PhC structure. The spectra of the QDs in GaAs slab show two main spectral peaks at long wavelength and one or two very weak spectral peaks at short wavelength (Figure 2). The spectra of the QDs in GaAs slab with PhCs split into multi-peak spectra, especially two relative strong peaks appeared in the short wavelength region. The spectra under different pump distances contain excitation and collection angle information. Because the incident angle of pump light and the collection angle of emission light vary with the pump distances. The parameters in Figure 3(b) show that the PL spectral central wavelength almost doesn't change with the pump distance, which means that the multi-peak wavelength doesn't change with the incident angle and the collection angle in some degree. Moreover, it indicates that the interaction between light and the QDs is still in weak coupling region.

The amplitudes of different spectral components are shown in Figure 3(d). As discussed before, the pump distance around 6 is the position corresponding to maximum PL intensity (approximate focus state), while the pump distance around 16 is corresponding to minimum PL intensity. The difference from PL intensity is mainly due to the difference in pump intensity and the collection angle. The ratio of the maximum PL intensity at position 6 to the minimum PL intensity at position 16 for each

component was calculated. The obtained ratio for A_3 component (short-wavelength) reaches to 199, while the ratios for A_1 component and A_2 components are 3 and 8, respectively. The ratio for A_3 component at short wavelength is much larger than those for A_1 and A_2 components at long wavelength. The components at long wavelength of A_1 and A_2 are from ground-state emission and the first excited state emission, while A_3 component at shorter wavelength is from the second excited-state emission, a high excited state emission. It means that the higher pump intensity will result in larger enhancement to the higher excited state emission than those to the ground-state emission or the lower excited-state emission. Generally, the pump intensity (electrical field) in PhCs slab is higher than that in blank material without PhCs.

1.3 The spectra pumped with 40×objective

The PL spectra from InAs/GaAs QDs with PhCs under different pump distances by 40×/0.6 objective lens are shown in Figure 4 (a) and (b). The spectram in focus state is fitted into multi-Gaussian function (Figure 4 (b)). Three peaks appeared in the spectra in Figure 4 (a), in which the center wavelength is 1 263 nm, 1 192 nm and 1 112 nm, respectively. In the focus position of the objective lens, four peaks were observed and the respective central wavelength is 1 263 nm, 1 192 nm, 1 135 nm, and 1 112 nm as shown in Figure 4(b). Compared with Figure 2(a) and 2(b), the main difference is that one shortest wavelength peak at 1 135 nm appears in the focus state, while the wavelength of the other two peaks at long wavelength are similar to those in Figure 2(a). The peak at 1 135nm wavelength and the peak at 1 192 nm wavelength are enhanced when the sample is in focus state. In the experiment, the PL intensity of the short-wavelength component is enhanced by PhCs more than that of the long-wavelength component. From comparison of the spectra at different pump distances, it can be found that the PL intensity of the short-wavelength component in the focus state is enhanced obviously. Compared the spectra of QDs with PhCs in Figure 2(c), the spectra of the shortest-wavelength peak in Figure 4 is broad. The only difference condition between Figure 2 and Figure 4 is the magnification of the objective, which will affect the incident angle of the pump laser and the collection angle of the emission.

The spectra of the QDs in different excitation positions were measured and analyzed. As the spectra are fitted into multi-Gaussian function, the fitted parameters of central wavelength and spectral line width in different position are shown in Figure 4(c) and 4(d). In PhCs area, the spectral peak wavelengths (at 1 161 nm, 1 191 nm and 1 132 nm) and the spectral widths for the three long-wavelength components almost don't change with the excitation position changing. However, the situation changes for the shortest wavelength component. It can be found that the central wavelength of the shortest wavelength x_{c4} fluctuates, which varies between 1 092 nm and 1 119 nm, and the corresponding spectral width is 104 nm. The spectral width of the shortest wavelength component fluctuates drastically, and it is much wider than the

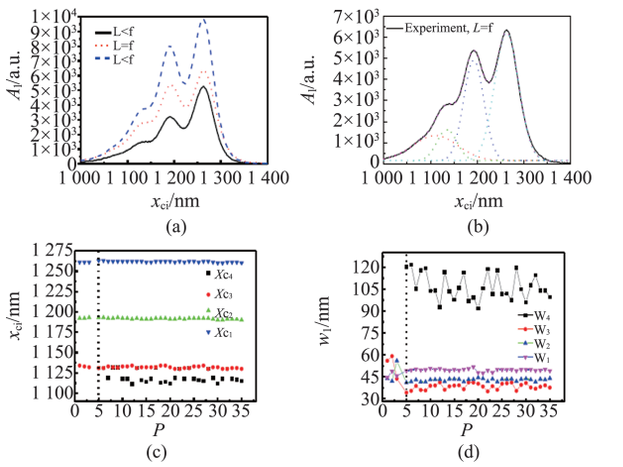

Fig. 4 The spectra of InAs/GaAs QDs with PhCs with different pump distance between the surface of sample and that of the objective of 40×/0.6. (a) The spectra of InAs/GaAs QDs with different pump distance in PhCs area. (b) The pumping distance was focal length (a). The (c) and (d) were the spectral parameters of QDs at different position (P) in the photonic crystals area with objective at focal position with 40×/0.6 objective lens. (c) Fitted central wavelength of the spectra in PhCs. (d) Fitted spectral width. Vertical dotted lines in both figures denote the boundary line for the area of no PhCs and that with PhCs.

图4 使用40×/0.6物镜测试不同距离下带有光子晶体的InAs/GaAs量子点材料的发光光谱.(a)光子晶体区域不同泵浦距离的测试光谱.(b)泵浦距离为焦距的测试光谱.量子点在光子晶体的不同区域的光谱拟合参数结果,测试条件均为在聚焦状态使用40×/0.6物镜进行测试.(c)光子晶体区域的中心波长的拟合结果.(d)光谱宽度的拟合结果.纵向点划线的位置区分了光子晶体区域和非光子晶体区域.

spectral widths of the other three peaks. Therefore, the shortest wavelength component may be caused by the emission of the wetting layer[4]. The spectral component at 1 132 nm, though appears in both blank and PhCs, the spectral width in PhCs is narrower than that in blank material. Moreover, the spectral widths of the first to the third spectral peaks (x_{c1} to x_{c3}) are much flatter varying with excitation positions in the PhCs' area in Figure 4 (c) than that in blank material. There are three wavelength peaks in the spectra of no-etched material (blank material). The spectral central wavelength and the spectral width of the first peak and the second peak at long wavelength maintain almost as constant when the pump position changes. Though the long three peaks have already appeared in the spectra without PhCs, they became stronger with PhCs, and the spectral wavelength and the spectral width didn't change with the excitation position and pump distance. These properties might have potential applications in space optical modulator and course wavelength division multiplex. In this study, the lattice constant of PhCs used was large, though we fabri-

cated it using holography method, actually, it can be made by conventional photo-lithography method, which is a very economical and practical way.

1.4 Results and analysis

Comparing the spectral parameters in blank and those in PhCs, the PL intensity of the short wavelength component is enhanced by PhCs compared to that of blank material without PhCs (Figure 5(a)). PhCs also caused the fluctuations of the central wavelength and the spectral width, which might be caused by the difference of the local density of the states in different position in PhCs. [10] Somewhere in PhCs, the spectra could not be fitted well into multi-Gaussian function, probably due to the asymmetry enhancement from the PhCs slab. As compared in Figure 5(a), the varying tendencies as function of wavelength of the enhancement factors are similar for both cases of 10×/0.25 objective and 40×/0.6 objective, while the enhancement factor for 40×/0.6 objective lens is a little higher than that for 10×/0.25 objective lens, and it is larger than 1 in the spectral range we measured. One reason of the enhancement to the QDs PL spectrum by PhCs is thought be due to the coupling between QDs emission and the leaky modes of PhCs slab of the GaAs/AlGaAs with QDs. The bandstructure is calculated and a part of it is shown in Figure 5(b), where the horizontal axis represents the wave vector (k), vertical axis is the normalized frequency (f) in unit of a/λ, where a is the lattice constant of the PhCs, and λ is the wavelength of light. At wavelength 1 100 nm corresponding normalized frequency 3.41 (a/λ) near which at Γ point, there are several modes, these modes are far above light line, so they are leaky modes. As the emission is coupled with these leaky modes, the PL of QDs will be enhanced. Therefore, the enhancement to the short-wavelength emission peaks in the spectra of QDs with PhCs is due to that the corresponding emission is resonant with the leaky modes of the PhCs slab. For another wavelength around 1 270 nm, the corresponding normalized frequency is 2.96 (a/λ), where the calculated modes at Γ point is spares (not shown). The enhancement of the PL intensity at 1 100 nm is as high as 7, while there is almost no enhancement of the PL intensity at 1270 nm. Similar to the explanation to the enhancement of the PL intensity by PhCs' bandstructure, the leak modes of the PhCs slab can also enhance the electrical field of the pump light[9]. The high pump intensity would enhance PL intensity of the excited-state emission and high excited-state emission (corresponding to the shorter wavelength spectra) due to state filling effect.

From the above analysis, the larger enhancement by PhCs to the short-wavelength region (high excited-state emission) than that to the long-wavelength region (ground and lower excited-state emission) results from two reasons: firstly, the emission in the short-wavelength region and the pump intensity are enhanced by PhCs. With high pump intensity, the high excited state could be excited and the PL efficiency was higher than those with low pump intensity due to state filling effect[11]. Secondly, the degeneracy of the excited sate is higher than

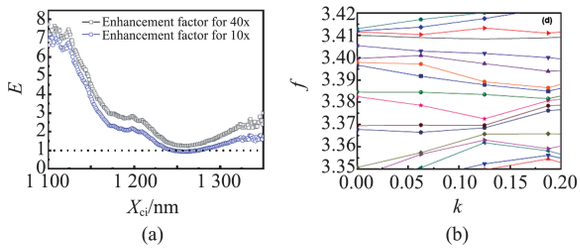

Fig. 5 (a) Comparison of the enhancement factors of PL with PhCs with 40 ×/0.6 and 10 x/0.25 objective lens at focal distance, $L=f$. (b) Calculated part of the bandstructrure of PhCs with the material in Figure 1(b) with ratio of 0.181 for radius of hole to lattice constant,where f is frequency and k is wave vector.

图5 (a)比较了光子晶体区域在40×/0.6和10×/0.25测试条件下聚焦状态的光致发光光谱的增益因子,L=f.(b)计算材料上制作的图1(b)中的光子晶体的能带结构,孔的半径和晶格常数比率是0.181,图(b)中f代表频率,k代表波矢

that of the ground state[5], which results in the higher PL efficiency of the excited state than that of the ground state. With enhanced pump intensity and the high degeneracy of excited state, the emission efficiency for the excited state and high excited state would increase more than that of the first excited state and that of the ground state. Therefore the enhancement to the high excited state emission by PhCs was larger than that to the lower excited state emission.

2 Conclusion

In conclusion, PhCs in GaAs/InAs QDs material were fabricated by holograph and wet etching method. The PL spectra in QDs material with PhCs were measured. The phenomena of multiple peaks of the PL spectra in different positions and pump distances in PhCs were analyzed by combining with the QDs characterization and the bandstructure of PhCs. The multi-peak in the spectra resulted from different order excited-state emission in PhCs area. The enhancement to the short-wavelength component in the spectra of InAs /GaAs QDs with PhCs was much larger than that to the long-wavelength component. The enhancement was regarded as the overlaps of the absorption of the pump light and the emission from QDs with leaky modes of PhCs slab, and the high degeneracy of the excited states of QDs. It could be found that PhCs could modify the PL spectral structure, and the high order excited-state emission could be significantly enhanced by PhCs.

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