Diffraction from excitonic diffraction grating

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Abstract. In this paper we consider two methods for the theoretical modelling of the reflection and diffraction spectra from an excitonic diffraction grating made from a quantum well with spatial modulation of the exciton resonance. The modelling was performed in the single-scattering approximation, and by the step-by-step approximations of the exact solution of Maxwell's equations. The expressions are obtained for the modulation of the inhomogeneous broadening of the exciton resonance. Theoretical results are compared with the experimental data for the quantum well grown on an ion irradiated substrate, and for a quantum well irradiated with ions after growth.

1. Introduction

A3B5 quantum wells are one of the promising media for creating functional elements for optical computing. The molecular beam epitaxy (MBE) makes it possible to produce high quality quantum wells (QWs) with the inhomogeneous broadening of the exciton resonance comparable to its radiative width [1,2]. In [3] and [4] we have proposed methods for creating the simplest resonant diffractive optical elements (DOEs) based on QWs — excitonic diffraction gratings. In [3] such gratings were created by the GaAs substrate ion beam irradiation prior to the MBE growth of QWs. In [4] the InGaAs/GaAs QWs were irradiated by the ion beam after the MBE growth [5]. In both cases, the modification led to the appearance of areas with an additional inhomogeneous broadening of the exciton resonance. Spatially-periodical modified areas allowed us to observe a new optical response from the sample — resonance diffraction.

A similar technological approach can be used to create other resonant diffraction elements based in operation principle on the traditional non-resonant elements, such as axicons [6], binary diffraction gratings able to control the phase and polarization of light [7], or coupling gratings [8], down to photonic crystals [9,10] and metamaterials [11].

To analyze the experimental data and calculate the new resonant diffractive optical elements, it is necessary to build a theoretical model describing the scattering of a plane electromagnetic wave on such a structure. In this paper, we compare two approaches — the single-scattering approximation and the step-by-step approximation of Maxwell's equations.

2. Setting up a problem

Consider the problem of light scattering on the structure shown in Fig. 1. The structure consists of two semi-infinite layers with refractive indices $n_1$ and $n_2$ ($n_2 > n_1$). A plane monochromatic wave is incident on the structure from the upper half-space at an angle $\theta$. The problem will be solved for the...
case of TM-polarized light incident at the Brewster angle ($\theta = \arctg(n_2/n_1)$) and small diffraction angles, which will allow us not to take into account interference phenomena associated with non-resonant reflection from the I/II interface. The exciton susceptibility of a quantum well located at a depth $h$ is spatially modulated along the $x$ axis. The growth axis is denoted as $z$. Let the QW layer susceptibility have the form of a periodic array of stripes of the same width:

$$g(x) = \begin{cases} g_1, x \in \left[mL, (m + \frac{1}{2})L\right] \\ g_2, x \in \left((m + \frac{1}{2})L, (m + 1)L\right), \end{cases}$$

(1)

where $L$ is the period of the spatial modulation, $g_1$ and $g_2$ are susceptibilities of odd and even stripes respectively, $m$ is the stripe number. The stripes susceptibilities are set as follows:

$$g_{1,2} = \frac{\Gamma_R}{\Delta \omega - i \Gamma_{NR1,2}},$$

(2)

where $\Delta \omega$ is the detuning of the incident light frequency from the exciton resonance frequency, $\Gamma_R$ is the radiative width of the exciton resonance, and $\Gamma_{NR1,2}$ are the nonradiative broadenings of odd and even stripes, respectively. So we will consider periodical spatial modulation of nonradiative broadening of the exciton resonance, which is realized in [3] and [4].

![Figure 1. Structure layout.](image)

When light is scattered on the structure, diffraction reflexes will appear in addition to the reflection, propagating at angles determined by the following formula:

$$\varphi = \arcsin \left(\sin \theta - \frac{n \lambda}{L}\right),$$

(3)

where $\lambda$ is the wavelength of the incident light, $n$ is the diffraction order ($n = 0$ corresponds to the reflection).

We wish to find the reflection coefficient $K_0$ and the diffraction efficiency of the first reflex $K_1$ (the fraction of the incident light intensity scattered in the corresponding directions). For this purpose, two different methods will be utilized in this work: the single scattering approximation and the step-by-step approximation of the exact solutions of the Maxwell's equations.

3. Single scattering approximation

We introduce the amplitude of the electric field of the incident plane wave:

$$E_{in}(\vec{r}, t) = A e^{-i\omega t} e^{i \vec{k} \cdot \vec{r}},$$

where $\vec{k}$ is the light wave vector, $\vec{r}$ is the radius-vector. We assume that as a result of the scattering, the light frequency $\omega$ does not change, and the factor $e^{-i\omega t}$ can be omitted. We will denote the scattered field $E_{out}$. The single scattering approximation can be expressed as the proportionality of the scattered field $E_{out}$ and the incident field $E_{in}$ in the QW layer. The coefficient of proportionality is the susceptibility:

$$E_{out}(x, z = h) = g(x)E_{in}(x, z = h),$$

(4)
We will search for the scattered field in the form of expansion in a Fourier series in x-projections of the wave vector (we denote this variable as $q$): 

$$E_{\text{out}}(x, z = 0) = A \int R(q) e^{i q x} d q.$$ 

Let's substitute this expression into (4), and calculate the inverse Fourier transform. Let $k_x$ denote the projection of the wave vector of the light incident on the sample: 

$$R(q) = \frac{1}{2\pi} \int g(x) e^{-i(q-k_x)x} dx.$$ 

We will substitute the Fourier transform for the case of a periodically modulated quantum well. The reflection coefficient and the diffraction efficiency can be found by isolation of the components that propagate at respective angles:

$$K_0 = \frac{\Gamma_R^2 (\Delta \omega^2 + \Gamma^2_{N_R})}{(\Delta \omega^2 + \Gamma_R^2)^2 + (\Delta \omega^2 + \Gamma_{N_R}^2)^2},$$

$$K_1 = \frac{4 \Gamma_R^2 (\Gamma_{N_R2} - \Gamma_{N_R1})^2 (\Delta \omega^2 + \Gamma_{N_R1}^2)}{(\Delta \omega^2 + \Gamma_R^2)^2 + (\Delta \omega^2 + \Gamma_{N_R}^2)^2},$$

For the case of absence of the spatial modulation ($\Gamma_{N_R1} = \Gamma_{N_R2} = \Gamma_{N_R}$) the diffraction efficiency vanishes ($K_1 = 0$), and reflection coefficient has the following simple form:

$$K_0 = \frac{\Gamma_R^2}{\Delta \omega^2 + \Gamma_{N_R}^2}.$$ 

This expression leads to an overestimation of the reflection coefficient $K_0$. Thus, for small $\Gamma_{N_R}$ near the resonance ($\Delta \omega \to 0$), the coefficient $K_0$ may take nonphysical values $K_0 > 1$. Thus, the single scattering approximation is applicable only $\Gamma_{N_R} >> \Gamma_R$.

4. Step-by-step approximation of Maxwell equations solution

The single scattering approximation is valid only in the case of a small radiative width of the exciton resonance $\Gamma_R$. An accurate expression can be obtained by solving the Maxwell’s equations. This problem has an exact analytical solution for the case of the reflection form a homogeneous quantum well ($g_1 = g_2$) [1]. For the case of an excitonic diffraction grating, an approximate solution can be obtained by an expansion with respect to a small parameter – the contrast of the spatial modulation. Below is an approximate solution obtained when taking into account the first two terms of this expansion:

$$K_0 = \frac{\Gamma_R^2 (\Delta \omega^2 + \Gamma_{N_R1}^2)^2}{(\Delta \omega^2 + \Gamma_{N_R1}^2)^2 + (\Delta \omega^2 + \Gamma_{N_R2}^2)^2},$$

$$K_1 = \frac{1}{\pi^2} \frac{4 \Gamma_R^2 (\Gamma_{N_R2} - \Gamma_{N_R1})^2 (\Delta \omega^2 + \Gamma_{N_R1}^2)}{(\Delta \omega^2 + \Gamma_{N_R1}^2)^2 + (\Delta \omega^2 + \Gamma_{N_R2}^2)^2}.$$

where $\tilde{\Gamma}_{1,2}$ are introduced as follows:

$$\tilde{\Gamma}_{1,2} = \sqrt{\frac{\Gamma_{N_R1}^2 + \Gamma_{N_R2}^2 + (\Gamma_{N_R1} + \Gamma_{N_R2} + \Gamma_R)^2}{2}}.$$ 

One could see that in the case of modulation absence ($\Gamma_{N_R1} = \Gamma_{N_R2} = \Gamma_{N_R}$) the diffraction efficiency $K_1$ is equal to zero, and $\tilde{\Gamma}_1 = \Gamma_R + \Gamma_{N_R}$, $\tilde{\Gamma}_2 = \Gamma_{N_R}$. In this case the reflection from the homogeneous quantum well could be obtained analogous to [1]:

$$K_0 = \frac{\Gamma_R^2}{\Delta \omega^2 + (\Gamma_R + \Gamma_{N_R})^2}.$$ 

5. Comparison of approximations and discussion

Figure 2 shows the reflection and diffraction spectra calculated by formulas (5) and (6) for typical values of exciton resonance parameters for a thin InGaAs/GaAs quantum well at liquid helium temperature ($\Gamma_R = 40 \mu eV$, $\Gamma_{N_R1} = 100 \mu eV$, $\Gamma_{N_R2} = 200 \mu eV$).
Figure 2. Reflection (a) and diffraction (b) spectra calculated in single scattering approximation (blue dashed curves) and step-by-step Maxwell’s equation solution approximation (red solid curves). Inset shows diffraction spectra in logarithmic scale.

As mentioned above, an approximate solution of Maxwell’s equations in the absence of spatial modulation leads to the Lorentz function, which is an exact solution for the uniform quantum well. In the case of spatial modulation, the qualitative behavior away from the resonance $K_0(\Delta \omega) \sim \frac{1}{\Delta \omega^2}$ is preserved. This behavior is also predicted by the single-scattering model, but it leads to an overestimation of the resonant reflection coefficient (Fig.1 a). This is due to the fact that in this model the radiative width does not contribute into the total width of the resonance. Such a difference is not significant for low-quality quantum wells or at elevated temperatures, but in high-quality samples at liquid helium temperatures, the radiative width and non-radiative broadening could be comparable [1, 12, 13, 14].

The diffraction reflex is formed due to the spatial modulation contrast, so it decreases with detuning much faster: $K_1(\Delta \omega) \sim \frac{1}{\Delta \omega^4}$. For the same reason, the half width at half maximum of the peak in the diffraction spectrum is smaller than that in the reflection spectrum.

Figure 3 shows a comparison of the theoretical fit and experimental diffraction spectra from different excitonic gratings for the cases of the ion irradiation of a quantum well after epitaxial growth (a) and of the ion irradiation of the GaAs substrate with the subsequent MBE growth of quantum wells (b).

Spectra shown in Fig. 3 (a) corresponds to the In$_{0.015}$Ga$_{0.985}$As/GaAs quantum well 4.5 nm thick with a 60 nm GaAs covering layer. The quantum well was irradiated by a 35 keV He$^+$ ion beam in Zeiss ORION helium ion microscope. Irradiated pattern was an periodic array of 400 nm and 150 mkm length rectangles with period 800 nm. Presented spectra correspond to the ion beam dose $5 \cdot 10^{11}$ cm$^{-2}$. The grating period is nearly equal to the resonant wavelength of the exciton resonance.

Spectra presented in Fig. 3 (b) corresponds to a quantum well grown on the pre-irradiated substrate. A GaAs wafer was irradiated by 30 keV Ga$^+$ ion beam using Carl Zeiss Crossbeam 1540XB workstation by 2 nA ion beam current. The irradiated pattern was a periodical array of 400 mkm length lines drawn with 9 mkm period. Ion beam dose was 0/1 nA s cm$^{-1}$. On the irradiated substrate several quantum wells were grown. Presented spectrum corresponds to a 2 nm thick In$_{0.02}$Ga$_{0.98}$As/GaAs quantum well separated by 150 nm buffer from the irradiated substrate. The ratio of the grating period to the wavelength of the exciton resonance is around 11:1.

More details of samples and measurements technique are given in [4] and [3] for Fig. 3 (a) and (b) respectively. In both cases, the diffraction efficiency far from the exciton resonance decreases more slowly than Gaussian ($e^{-\Delta \omega^2}$), but faster than Lorentzian ($\sim \frac{1}{\Delta \omega^2}$), and is well described by the theoretically predicted dependence $K_1(\Delta \omega) \sim \frac{1}{\Delta \omega^4}$. 
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Figure 3. Comparison of the theoretical fit of the diffraction spectra by formula (5) (red dashed curve), Lorentzian (green dashed) and Gaussian (dark red dashed) curves for an excitonic diffraction grating fabricated by the ion irradiation after MBE growth (a) and the ion irradiation of the GaAs substrate prior to the growth (b).

6. Conclusion
The paper compares two theoretical models for describing the resonant diffraction by an excitonic diffraction grating. The expressions obtained qualitatively describe the observed experimental diffraction spectra. The advantages of the single-scattering method include the ability to calculate diffraction by simply calculating the Fourier transform for an arbitrary grating profile. This model also makes it possible to qualitatively describe the decrease of the reflection and diffraction spectra with detuning. However, for sufficiently high-quality quantum wells at low temperatures, this model overestimates the value of the reflection coefficient and underestimates the diffraction efficiency. In this case, it is advisable to use an approximate solution of the Maxwell equation.

7. References
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