Light propagation through conventional and extreme-2D van-der-Waals resonant photonic crystals

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Abstract. We report on theoretical studies of short optical pulse propagation through resonant photonic crystals (RPCs) based on conventional II-VI quantum wells (QWs) and van-der-Waals (vdW) Bragg heterostructures composed of extreme-2D monolayers. The parameters needed for modelling were determined by fitting of the experimental reflectance spectra. The delay of the transmitted picosecond optical pulses is predicted to be larger in vdW structures than in the structures with QWs due to the higher radiative width of the exciton resonance. Moreover, in the vdW RPCs, the pulse shape undergoes smaller modification owing to the almost linear dispersion of the “slow” mode. High-quality extreme-2D monolayers will allow for even stronger delays without distortion and small intensity decrease, needed for nanophotonics applications.

1. Introduction

Quantum information processing has been playing an important role in the world since quantum computers came into sight. Therefore, the study of the light pulse propagation through structures becomes crucial. The optical pulse can change its shape and slow down while travelling through crystals. Slowing down of light frequently exploits a sharp change in the refractive index near a resonance, which results in the decrease of a group velocity [1]. The refractive index corresponding to “slow” modes in photonic crystals can be increased by dozens of times. Yet this approach often ties with a strong attenuation of the pulse. One of the promising methods is to use optical modes in RPCs, where exciton resonances coincide in frequency with optical modes [2, 3]. In the RPCs, light dispersion is strongly modified near the Bragg frequencies that influences the slow mode performance. Previous studies experimentally demonstrated that RPC with a simple unit cell can provide the delay up to half of the incoming pulse width with attenuation of the intensity up to 1% [4]. We have shown that varying the parameters of the QWs and the design of the unit cell one can control the attenuation of light, its shape change and the group velocity [5, 6]. In such structures, slight detuning of exciton resonance from Bragg condition can provide a smooth control of the dispersion law of the “slow” modes. Hence, we can achieve light velocities as low as \( v = c/50 \) with only 3-5 times attenuation.

In this paper, we investigate a novel type of RPCs – van-der-Waals (vdW) Bragg structures. They consist of extreme two-dimensional (2D) vdW monolayers, e.g., transition metal dichalcogenides WSe2, MoS2, etc. Such extreme-2D materials have begun to draw attention since the appearance of graphene [7]. Heterostructures based on the extreme-2D monolayers offer a unique opportunity to design nanodevices with desired optical properties. We present a comparative study of light propagation through the conventional RPCs based on QWs and through vdW heterostructures comprising stacks of 2D monolayers, whose position in a dielectric matrix (SiO2) corresponds to the Bragg condition.

2. Structures and parameters
To model light propagating through RPCs we need the material parameters such as the exciton energy (ℏ\(\omega_0\)), the radiative width (\(\Gamma_0\)) and the non-radiative width (\(\Gamma\)) of the exciton resonance. These parameters can be obtained by fitting the experimental reflectance spectra with theoretical equations obtained using the transfer matrix technique. In this approach, each layer in a structure, i.e., a quantum well or a barrier, is described by a transfer matrix relating the amplitudes of the electric field at the left and right edges of the layer. The transfer matrix through several layers is the product of the transfer matrices of these layers. As a result, the dispersion law, the reflection and transmission coefficients can be calculated from the matrix equation. The reflectance from the structure comprising a 2D layer or a QW, a substrate and a coating layer reads:

\[
R = \frac{|r_c + \frac{\Gamma_0}{\omega - \omega_0 + i(\Gamma_0 + \Gamma)}|}{2}
\] (1)

The term \(r_c\) in the right-hand-side of the Eq. (1) describes Fresnel reflection of the incoming light from the substrate and the coating layer. It depends on the width of layers and dielectric contrast between them. The second term in Eq. (1) shows the reflection associated with exciton resonance in the 2D layer or QW. Multiple light reflections are neglected in Eq. (1), while taken into account natively in the transfer matrix simulation.

Figure 1. (a) Structure of the extreme 2D WSe\(_2\) crystal on a substrate. (b) Experimental reflectance spectra of WSe\(_2\) (black line, from Ref. [9]) and its fit by Eq. (1) (red line). (c) A sketch of vdW RPC.

As a model system for the conventional RPCs we investigated the spectra of the ZnSe/ZnMgSSe structure with a single quantum well grown on GaAs epitaxial buffer layer pseudomorphically to GaAs (100) substrate by molecular beam epitaxy (MBE) [8]. Exciton properties turned out to be \(\hbar \omega_0=2.83\) eV, \(\hbar \Gamma_0\sim0.17\) meV, and \(\hbar \Gamma\sim1.9\) meV. For vdW heterostructures, we have taken the reflection spectra measured at the low temperature (4 K) in a monolayer of the transition metal dichalcogenide WSe\(_2\), deposited on top of SiO\(_2\) layer on Si substrate (Figure 1a), reported in [9]. Our fit of the experimental spectrum is shown in Figure 1b. The determined parameters of the exciton resonance are \(\hbar \omega_0=1.75\) eV, \(\hbar \Gamma_0\sim0.3\) meV and \(\hbar \Gamma\sim3.0\) meV. The large value of the non-radiative exciton width is due to
inhomogeneous broadening of the exciton resonance. The obtained parameters correspond to the typical extreme 2D structures of moderate quality. We should notice that radiative and non-radiative parameters of the exciton resonances for extreme-2D vdW monolayers can be obtained from the other reflectance experiments [10, 11]. The variation of parameters indicates that 2D crystals are still not mature in terms of fabrication.

Knowledge of the parameters of the exciton resonances can allow us to model pulse propagation through RPC Bragg structures. Our conventional RPCs consist of ZnSe quantum wells located between ZnMgSSe barriers of a finite width. In vdW RPCs, monolayers of WSe$_2$ are separated by SiO$_2$ spacers, see Figure 1c. Note that background reflection of the monolayer cannot be neglected. While the thickness of the monolayer is small, dielectric contrast with spacer is strong. The period of both conventional and vdW RPCs is chosen in such way that Bragg condition is fulfilled at the frequency close to exciton resonance.

3. Results and discussions
Using the transfer matrix technique, described in detail in [6], we have calculated the light dispersion inside the vdW and conventional RPCs and modelled pulse transmittance through the structures with 70 resonant layers, see Figure 2. We considered two values of the non-radiative exciton resonance width, $\hbar \Gamma = 0.3$ meV (red line) and $\hbar \Gamma = 3.0$ meV (black lines), while keeping the parameter $\hbar \Gamma_0 = 0.3$ meV fixed for both types of RPCs. Incident pulses had the carrier frequency slightly detuned from the exciton resonance ensuring the balance between pulse delay and attenuation.

Figure 2. The light dispersion and transmitted pulse envelope for the conventional RPC (a, c) and the extreme-2D vdW RPC (b, d) consisting of 70 layers. For both RPCs, the non-radiative exciton width is
chosen $\hbar\Gamma=0.3$ meV (red line) or $\hbar\Gamma=3.0$ meV (black line), $\hbar\Gamma_0=0.3$ meV. The initial pulse envelope is a Gaussian with 2 ps half width, see the dotted lines.

Comparing of the results for the conventional and the vdW RPCs we conclude that vdW 2D Bragg structure has an advantage in the light delay over the conventional II-VI RPCs. The pulse delay is about 2 ps for the vdW RPC as compared to 1 ps for the conventional RPC. Such delay is comparable with the initial pulse half width (2 ps). On the other hand, transmitted pulse intensity in the conventional PRC is 1.5 times larger than in vdW structures. We note that the increase of structure quality (i.e., decrease of the non-radiative exciton broadening) will lead to the improvement in the pulse delay and attenuation, see red lines in Figure. 2d. Interestingly, light dispersion of the vdW RPC reveals the almost linear behavior of the “slow” mode throughout the half width of the stopband. This ensures minimal distortion of the pulse. Contrary, in the conventional RPCs, the linear behavior is not traceable.

Finally, the Bragg structure consisting of 20 periods ZnSe (9.7nm)/ZnMgSSe (64.4nm) has been grown by MBE on GaAs (001) substrate via 0.2 $\mu$m-GaAs buffer layer using double-chambers MBE setup. The total thickness of the structure was 1.51 $\mu$m. Modelling of the reflection spectra shows that peculiarities related to interaction of exciton and Bragg resonances are very small for a such thin RPC Bragg structure. However, the exciton resonance was clearly observed in the reflection experiments. The MBE growth of the 70-100 periods ZnSe/ZnMgSSe RPC Bragg structure with the total thickness of 5-7 $\mu$m looks as a rather complicated task because of the strong requirements for both growth temperature and Se flux stability during the growth run. In case of MBE growth of S-containing alloys the time drift of both parameters results in growth rate (and, in turn, in thickness) and sulfur content variations throughout the RPC structure [12]. We hope that better quality and longer length of the RPCs will find someday various applications in nanophotonics.

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