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Optical spectra and inhomogeneous broadening in CdTe/CdZnTe MQW structures with defects

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Abstract
Optical spectra of Bragg multiple quantum wells with defects are studied analytically and numerically. It is shown that in systems with relatively strong exciton–photon coupling several different types of spectrum can be observed. The effects due to inhomogeneous exciton broadening are studied using numerical simulations.

1. Introduction

Optical properties of multiple quantum wells (MQWs) have attracted a great deal of interest recently [1–7]. Unlike other types of superlattice, excitons in MQWs are confined in the planes of the respective wells, which are separated by relatively thick barriers. Therefore, the only coupling between different wells is provided by the radiative optical field. The coupling results in MQW polaritons—coherently coupled quasi-stationary excitations of quantum well (QW) excitons and transverse electromagnetic field. The spectrum of short MQW structures consists of a number of quasi-stationary (radiative) modes with finite lifetimes. This spectrum is conveniently described in terms of super- or sub-radiant modes [1, 3]. When the number of wells in the structure grows, the lifetime of the former decreases, and the lifetime of the latter increases. In longer MQW structures, however, this approach becomes misleading, as discussed in [8], and a more appropriate description is obtained in terms of stationary modes of an infinite periodic structure. The spectrum of MQW polaritons in this case consists of two branches separated by a gap with a width proportional to the exciton–light coupling constant, Γ [1, 3]. If one is interested in properties of the gap region, a 'long system' usually means that it is longer than the penetration length of the radiation into the sample. The penetration length depends upon the frequency, therefore the system can be long enough for frequencies close to the centre of the gap, and still 'short' for frequencies in the vicinity of the band edges.

In a number of papers [2, 4, 6, 7] it was shown that the width of the polariton bandgap can be significantly increased by tuning the interwell spacing, a, to the Bragg condition, $a = \lambda_0/2$, where λ_0 is the wavelength of the light at the exciton frequency Ω_0. When a approaches λ_0, one of the Bragg bandgaps moves toward the polariton bandgaps simultaneously increasing in width. When $a = \lambda_0/2$, boundaries of two adjacent gaps become degenerate, and one wide gap with width proportional to $\sqrt{\Gamma\Omega_0}$ is formed. Detuning of the lattice constant from the exact Bragg condition removes the degeneracy and gives rise to a conduction band in the centre of the Bragg gap [8]. A well pronounced Bragg polariton gap was observed in recent experiments [7] with GaInAs/GaAs Bragg structures with the number of wells up to 100. Polariton effects arising as a result of this coupling open up new opportunities for manipulating optical properties of quantum heterostructures.

One such opportunity is associated with introducing defects in MQW structures. These defects can be either QWs of different compositions replacing one or several 'host' wells, or locally altered spacing between elements of the structure. It was shown in [9] that the defects in an infinite MQW structure can give rise to local exciton–polariton modes with frequencies inside the polariton gap. One should clearly distinguish these defect polariton modes from well known interface modes in layered systems or non-radiative two-dimensional polariton modes in ideal MQWs [10–12]. The latter exist only with the in-plane wavenumbers, $k_{||}$, exceeding certain critical values, while the local mode in a defect MQW structure exists at $k_{||} = 0$ and can be excited even at normal incidence. The detailed study of the local polaritons for different types of defect in Bragg MQWs was carried out in our recent papers [13, 14]. In these papers we obtained dispersion equations of local polaritons and analysed the dependence of the respective frequencies upon parameters of the system. In particular, we showed that a single defect in such structures gives rise to two local states, one below and one above the exciton frequency. In [13, 14] we also considered modifications of the optical spectra due to the local polaritons. Both exciton inhomogeneous and homogeneous broadening were treated with a single parameter of the non-radiative width within the framework of the linear dispersion theory [22]. Using GaAs/AlGaAs and GaInAs/GaAs systems for numerical examples we found that in the presence of both homogeneous and inhomogeneous broadening only defects produced by a local change of the interwell distance can induce significant modification of the spectra.

In this paper we focus on the effects of inhomogeneous broadening on the optical spectra of the defect MQWs. The role of inhomogeneous broadening in regular (without defects) MQW structures has been intensively studied (see, for instance, [15–17] and references therein). Usually two types of disorder, vertical and horizontal, are considered separately. The horizontal disorder refers to inhomogeneities, which exist within a single well, while the term vertical disorder describes random fluctuations between properties of different wells in structures with several QWs [16].

The objective of this paper is to assess the influence of the vertical disorder on defect-induced features in the optical spectra of defect MQWs. To this end we numerically construct a system of MQWs with a single defect allowing for exciton frequency to fluctuate from well to well. Using a computational approach based upon a blend of the transfer matrix method and the invariant embedding method, which was described in our paper [20], we calculate the optical spectra for several different realizations of our structure with consecutive averaging over these realizations. The numerical method developed in [20] is advantageous for calculations in the frequency region of bandgaps since it remains very stable even for extremely small values of the transmission coefficient. The numerical calculations are compared with the results of the linear dispersion theory. The comparison allows us to see how so-called motional narrowing of polaritons affects defect-induced features of the spectra. The motional narrowing describes partial averaging out of the disorder due to coherent coupling between wells in MQW structures and was studied theoretically [16] and experimentally [18] for regular MQW structures. Our calculations demonstrate manifestations of this effect in defect MQWs. As a model system for our calculations we use CdTe/CdZnTe MQWs studied experimentally in [19]. The advantage of this system over GaAs/AlGaAs and GaInAs/GaAs structures, which we considered in our previous papers, is much stronger exciton–photon coupling combined with a relatively moderate inhomogeneous broadening. We show that owing to these properties CdTe/CdZnTe structures demonstrate a greater variety of spectra with more pronounced effects of motional narrowing.

2. Optical spectra in ideal MQWs with defects

In order to describe optical properties of QWs one has to take into account the coupling between retarded electromagnetic waves and excitons. This is usually done with the use of the non-local susceptibility determined by energies and wavefunctions of a QW exciton [1, 12]. The treatment of the exciton subsystem can be significantly simplified if the interwell spacing is much larger than the size of a well itself. In CdTe/CdZnTe MQW structures studied in [19], on which we base our numerical examples, the width of the QW layer amounts only to about 10% of the period of the structure. In this case, one can neglect the overlap of the exciton wavefunctions from neighbouring wells and assume that an interaction between well excitons occurs only due to coupling to the light. It is also important that the width of the wells is also considerably smaller than the exciton's Bohr radius, and, therefore, one can neglect the spatial extent of the wells, and describe them with the polarization density of the form $\boldsymbol{P}(\boldsymbol{r}, z) = \boldsymbol{P}_n(\boldsymbol{r})\delta(z - z_n)$, where $\boldsymbol{r}$ is the in-plane position vector, z_n represents the coordinate of the nth well and $\boldsymbol{P}_n$ is the surface polarization density of the respective well. Optical response of QW excitons with homogeneous broadening can be successfully described by the linear dispersion theory (see, for instance, [22]), based upon a single oscillator phenomenological expression for the exciton susceptibility χ, which can be defined as a coefficient of proportionality between the electric field, E_n at the nth well and respective surface polarization density P_n: $\chi = c\Gamma_n/\pi(\Omega_0^2 - \omega^2 - 2i\gamma\omega)$, where Ω_0 and γ are the 1s exciton frequency and the relaxation parameter, respectively, and Γ_n is a radiative light time of a single QW, which characterizes the strength of the light–exciton coupling.

In an infinite pure system, $\Gamma_n = \Gamma_0$, $\Omega_n = \Omega_0$ and $z_n = na$, where $a = \lambda_0/2$ is the Bragg interwell separation. The spectrum of ideal periodic MQWs with broadening parameter $\gamma = 0$ has been studied in many papers [1,3,8,21,23,24]. In the specific case of Bragg structures, the exciton resonance frequency, Ω_0, is at the centre of the bandgap determined by the inequality $\omega_l < \omega < \omega_u$, where $\omega_l = \Omega_0\left(1 - \sqrt{2\Gamma_0/\pi\Omega_0}\right)$ and $\omega_u = \Omega_0\left(1 + \sqrt{2\Gamma_0/\pi\Omega_0}\right)$ [8]. In the formed bandgap the electromagnetic waves decay with the penetration (localization) length, which has a minimum value,

$$l_{\text{loc}} \approx a\sqrt{\frac{\Omega_0}{2\pi\Gamma_0}}, \tag{1}$$

at the centre of the gap. This quantity characterizes the depth of the optical barrier presented by the gap, and one can see from equation (1) that an increase in the exciton–photon coupling deepens the barrier. Using data available from the literature, one can estimate the minimum number of wells in MQWs required to fully observe effects discussed in this paper; we found that this number is about 70 for GaAs/AlGaAs [4] or 100 for GaInAs/GaAs, [7] but can be as small as 50 for CdTe/ZnCdTe [19]. We shall see, however, that the defect-induced effects can be observed even in shorter structures.

In this paper, we consider three types of defect. First of all, one can replace an original QW with a QW with different exciton frequency Ω_1 (Ω-defect). This can be experimentally achieved by varying the composition of the semiconductor in the well. Another possibility is to perturb an interwell spacing between a pair of wells (a-defect), and, finally, we consider a defect formed by changing the widths of two adjacent barriers in a way which keeps their sum equal to the double width of the unperturbed structure (b-defect). Experimental realization of the last two defects is simple and can be done at the sample growth stage. The eigenfrequencies of the local polariton states associated with each of the defects were studied in [13,14], and in this paper we focus upon optical spectra of these structures. Neglecting exciton broadenings, expressions for transmission coefficients were also obtained in [14]. It turns out that while all three defects can lead to the resonant transmission of light through the structure, they result in qualitatively different spectra. We showed in [14] that in the case of the Ω-defect only one of the two local modes, with the eigenfrequency, $\omega_{\rm def}$, lying not too close to the boundaries of the polariton gap or the centre of it, significantly contributes to the optical spectra. In the vicinity of the local mode, the transmission coefficient can be approximately presented in the following form (assuming that the defect layer is located in the centre of the structure) [14]

$$T = \frac{4\gamma_\Omega^2}{4\gamma_\Omega^2 + Q^2} \frac{\left(\omega - \omega_T + \frac{4\gamma_\Omega^2}{Q} + Q\right)^2}{\left(\omega - \omega_T + \frac{4\gamma_\Omega^2}{Q}\right)^2 + 4\gamma_\Omega^2}. \tag{2}$$

The transmission spectrum described by equation (2) has a shape known as a Fano resonance, where ω_T is the resonance frequency, which is radiatively shifted from Ω_1, and at which the transmission turns to unity. The parameters γ_Ω and $Q = \omega_T - \Omega_1$ describe the width and the asymmetry of the resonance respectively. The width parameter, γ_Ω, exponentially depends upon the length of the system according to

$$\gamma_\Omega = \pi\Omega_0 \left(\frac{\omega_{\rm def} - \Omega_0}{\Omega_0}\right)^2 {\rm e}^{-\kappa Na}, \tag{3}$$

where κ is the inverse penetration length at the local frequency. At $\omega = \omega_T - Q = \Omega_1$ the transmission equals zero, which is a signature of Fano resonances. An actual possibility to observe the Fano resonance in the considered situation strongly depends on the strength of absorption in the system, which must be at least smaller than Q. More detailed discussion of absorption-related effects is given below.

In the case of the a- and b-defects, both local frequencies are located not too close to the gap centre or to the gap boundaries [14], and one could expect the resonance transmission to occur at two frequencies—one above Ω_0, and the other below. Expending the transmission coefficient in the vicinity of one of the local modes one obtains a symmetric Wigner–Breit type of spectrum for both types of defect, if the defect is placed in the centre of the structure. The half-width, γ_a, of the resonance Lorentzian for the a-defect is given by

$$\gamma_a = 2\frac{(\omega_{\rm def} - \Omega_0)^2(\omega_{\rm def} - \omega_l)^{3/2}(\omega_u - \omega_{\rm def})^{3/2}}{(\omega_u - \Omega_0)^4}{\rm e}^{-\kappa Na}, \tag{4}$$

and the respective quantity for the b-defect is

$$\gamma_b = 2\frac{(\omega_{\rm def} - \Omega_0)^2}{\omega_u - \Omega_0}{\rm e}^{-\kappa Na}. \tag{5}$$

The frequencies ω_T, where the transmission coefficients have the maximum value of unity, are determined by the parameters of the defects [13, 14]. The order of magnitude of the pre-exponential factors in equations (3)–(5) depends upon the closeness of the frequencies of the respective local modes to the centre of the gap, Ω_0. In the case of the Ω-defect, $\omega_{\rm def}$ is mostly determined by the exciton resonance of the defect layer, Ω_1 [13, 14]. For the a-defect, $\omega_{\rm def} - \Omega_0 \propto \sqrt{\Gamma\Omega_0}$, which is a relatively large quantity, so one could expect to find in transmission spectra of the system with the a-defect two broad maxima well separated from the gap centre. In the case of the b-defect, $\omega_{\rm def} - \Omega_0 \propto \Gamma^{3/4}\Omega_0^{1/4}$ is much smaller than the respective quantity for the a-defect. Therefore, optical spectra of the structures with the b-defect can have two narrow peaks located relatively close to the gap centre. We see that different defects result in local states having different positions relative to the centre and the boundaries of the gap, and that this difference affects significantly the width of the respective resonances.

3. The role of homogeneous and inhomogeneous broadening

The location and the width of the resonances of ideal structures determine the shape of the spectra in real materials. In real systems, the resonance enhancement of the transmission is usually limited by homogeneous and inhomogeneous broadening of exciton resonances. Two cases are possible when exciton damping is taken into account. It can suppress the resonance transmission, and the presence of the local states can only be observed in relatively short systems as a small enhancement of absorption at the local frequency. External radiation in this case is not effectively coupled with the modes inside the system and is mostly reflected. In the opposite case, when the resonance transmission persists in the presence of damping, the coupling between inside and outside modes is strong, and one can expect enhancements in both transmission and absorption spectra. In this case, there is a coherent coupling between the exciton and the electromagnetic field, so that the local states can be suitably called local polaritons. Qualitatively we can assess the effect of absorption on resonances by looking at the widths of the respective spectra. These widths exponentially decrease with the length of the system; consequently, in sufficiently long systems all the resonances disappear. However, pre-exponential factors make different defects behave differently at intermediate distances. A simple qualitative estimate would require that the width of the resonances be smaller than the exciton relaxation parameter. Therefore, the resonances, where the pre-exponential factor of the width is considerably larger than the relaxation parameter, can be observed in systems of the intermediate length. On the one hand, the length must be greater than the localization length of the respective local mode. On the other hand, it must be small enough for the width of the resonance to remain larger than the exciton relaxation parameter. The Fano resonance arising in the case of the Ω-defect, though, requires special consideration since its vitality depends on the asymmetry parameter Q rather than upon the width parameter γ_Ω. Although the latter is determined by the large pre-exponent (of the order of the exciton resonance frequency Ω_0), the former is of the order of the light–exciton coupling constant Γ_0, which is much smaller. The survival of the Fano resonances is, therefore, determined by the ratio of the relaxation rate to the asymmetry parameter Q.

The typical value of the rate of the exciton relaxation (homogeneous broadening) in semiconductor QWs is of the order of tens of μeV (its accurate value is known only for a few systems, for which time-resolved spectra are available; see, for instance, [4] for the data on GaAs/AlGaAs MQWs).

The radiative rate varies within a wide range of magnitudes from relatively small values of tens of μeV for GaAs-based structures (27 μeV for InGaAs/GaAs MQW studied in [7]) to hundreds of μeV in CdTe-based structures, and to even larger values in nitride-based MQWs. If only the homogeneous broadening were present, we could immediately conclude that the Fano resonance in MQWs with Ω-defects could not exist in GaAs-based structures, but may survive in systems with stronger light–exciton coupling. As far as the a-defect is concerned, the pre-exponent of the resonance width in this case is of the order of magnitude of the gap width, which is proportional to $\sqrt{\Gamma_0\Omega_0}$. It is considerably larger than total non-radiative exciton broadening (which includes both homogeneous and non-homogeneous mechanisms) for all the systems in which the polariton gap can be in principle observed. Optical spectra of MQW systems with this defect were considered in [14] within the linear dispersion theory. Numerical simulations carried out by the method outlined above showed that inhomogeneous broadening practically does not affect the spectra, therefore, for MQWs with the a-defect one can use results obtained in [14] with only homogeneous broadening taken into account. For the b-defect the pre-exponential factor in the respective resonance width is of the order of Γ_0, and the conclusions we drew regarding the Ω-defect apply here as well.

Inhomogeneous broadening is usually significantly larger than the homogeneous one and plays an important role in the formation of the spectra. For the vertical disorder (for systems with an infinite exciton mass there is no difference between horizontal and vertical disorder as we discussed above), the role of the inhomogeneous broadening can be described in terms of the destruction of the coherence between different wells. For Bragg MQWs with defects, the role of the disorder can be qualitatively explained in more specific terms. As shown in [8], small deviations of an ideal structure from the Bragg condition result in a narrow conduction band appearing in the centre of the polariton gap. These deviations, therefore, would promote transmission (and absorption) at the exciton frequency. Now, if a local state due to a defect in a Bragg structure fell within this transmission window, it would be washed out when deviations from the Bragg conditions are introduced. At the same time the local states, which would arise in the Bragg structure farther away from the centre, are not significantly affected by small deviations from the Bragg condition. Random deviations of the exciton frequency from the Bragg value create local transmission windows, which, however, do not lead to the global enhancement of the transmission through the entire system because of the homogeneous broadening. But it does lead to the enhancement of the absorption in the centre of the gap with the width of the absorption maximum approximately equal to the width of the distribution of the exciton frequencies. One can suggest, therefore, that the inhomogeneous broadening mostly affects only the part of the spectra within this window around the exciton frequency. The local states which would arise in an ideal system that are close to Ω_0 will not survive the inhomogeneous broadening, while the states with frequencies farther away will not be affected by the disorder in any significant way. Because of the strong exciton–photon coupling in CdTe-based structures, the local states due to the b-defect would arise in such systems far enough from Ω_0, and they would be wide enough to survive both inhomogeneous and homogeneous broadening.

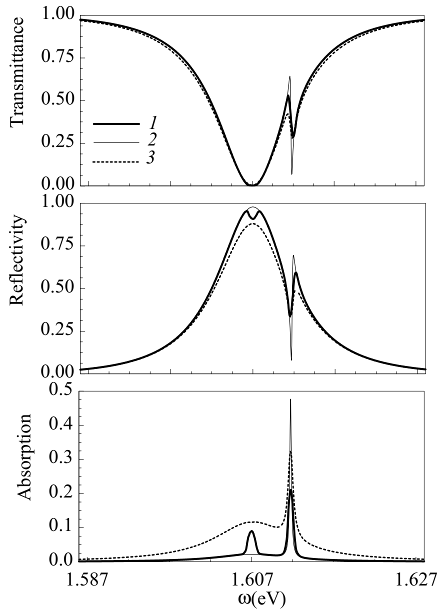

Figure 1. Transmission, reflection and absorption coefficients for the Ω-type defect. The defect is placed in the centre of the MQW with 41 QWs. The exciton frequency of the defect well Ω_1 is chosen such that $(\Omega_1 - \Omega_0)/\Omega_0 = 1.003$. Numerical values of all other parameters were taken for CdTe/CdZnTe structures studied in [19]. The bold solid curve shows results obtained using numerical Monte Carlo simulations, the thin solid curve shows spectra with inhomogeneous broadening neglected and the dashed curve presents the results of the linear dispersion theory with inhomogeneous broadening combined with the homogeneous one in a single parameter.

We complement the qualitative arguments presented above with numerical simulations of the optical spectra based upon the Monte Carlo approach described above. We use parameters of the CdTe/CdZnTe system [19] (Ω_0 = 1.607 eV, Γ = 0.12 meV, total non-radiative broadening, $\gamma_{\text{hom}} + \gamma_{\text{inhom}}$ = 0.3 meV) for our computations. In the absence of time-resolved measurements for this system, we could not separate contributions from homogeneous and non-homogeneous widths. Therefore, we assume that most of the total non-radiative width comes from the inhomogeneous broadening, which is true for most of the known QW systems. The exact value of the homogeneous contribution is not then very important, and we partition the sum rather arbitrarily using typical values for the homogeneous width in semiconductor QWs as a guide. We assume that γ_{hom} = 0.05 meV and $\gamma_{\text{inhom}} = 0.25$ meV. The results of calculations are presented in figures 1 and 2.

Figure 1 depicts transmission, reflection and absorption spectra of an MQW system consisting of 40 periods of CdTe/CdZnTe MQWs with an Ω_{def}. The defect frequency was chosen equal to Ω_1 = 1.612 eV. The curves in these figures represent the results of the Monte Carlo simulations (curve 1), calculations with only homogeneous broadening included (curve 2) and the spectra obtained within the linear dispersion theory, when all broadening

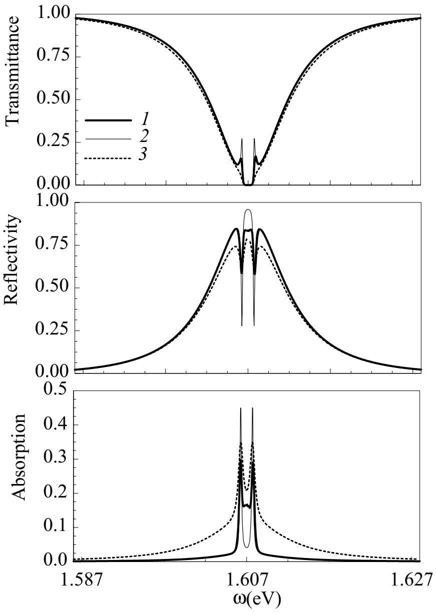

Figure 2. Transmission, reflection and absorption coefficients for the b-type defect in the centre of the MQW with 41 QWs. The widths of the defect barriers are $1.5a$ and $0.5a$. All other parameters are the same as for figure 1.

is treated effectively with a single constant (curve 3). These plots demonstrate a typical Fano-like behaviour in transmission and reflection spectra. Accounting for the inhomogeneous broadening reduces the amplitude of the respective maximum and minimum, but does not destroy the resonance, as expected on the bases of qualitative arguments given above. The difference between two different ways of including disorder (Monte Carlo simulations and the linear dispersion theory) is not very large in this case, though the linear dispersion theory underestimates the maximum of the transmission and leads to broader resonances. The narrowing of the spectra when disorder is treated correctly can be interpreted as a manifestation of the motional narrowing, but the effect is very weak in this case. The most prominent effects of the disorder are the dip in the reflection at the exciton frequency accompanied by the respective increase in absorption. These effects are manifestations of the local transmission windows discussed above, and are not related to the presence of local polaritons. As far as defect-related effects are concerned, the absorption spectrum demonstrates the most significant influence of the inhomogeneous broadening, which significantly reduces the peak value of the absorption, even compared with the linear dispersion theory in the region of the local polariton. Observation of the Fano resonance in defect MQWs provides a new powerful method of measuring characteristics of QWs. Positions of the maximum and minimum, their heights and widths give independent information on the exciton frequency, exciton–photon coupling, homogeneous and inhomogeneous broadening. The latter fact is particularly important because it allows one to measure parameters of the two mechanisms of broadening using CW experiments instead of more complicated time-resolved measurements.

Figure 2 presents the spectra for the b-defect. The widths of the two defect barriers were chosen to be $1.5a$ and $0.5a$, respectively, such that the distance between two affected wells remained $2a$. The frequencies of local states are much closer to the centre of the gap in this case than for the Ω-defect, and the spectral resonances are respectively much narrower. The effect of the motional narrowing is very pronounced in the transmission spectrum. While the linear dispersion theory predicts that the transmission resonances do not survive, the Monte Carlo simulations show clear peaks in the transmission. The absorption and reflection lines are significantly less broadened in the Monte Carlo approach. They basically follow the spectrum obtained without the inhomogeneous broadening, and the only apparent effect of the disorder is a reduction of the peak absorption. It is also interesting that the presence of the local states significantly alters the manifestation of disorder in the immediate vicinity of Ω_0—the disorder-induced dip/peak in the reflection/absorption is remarkably flattened at the level intermediate between the linear dispersion theory results and the calculations without the inhomogeneous broadening. Narrow lines in optical spectra of the systems with b-defects have a potential for applications in optical switches and modulators. The contrast in, for example, reflectivity can be increased by creating several such defects in a structure, and then a small shift in the exciton frequency using electric field can move the system from the high-reflectivity to the low-reflectivity state.

Acknowledgments

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