Polariton effects in multiple-quantum-well structures of $CdTe/Cd_{1-x}Zn_xTe$

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Strong effects of coupling between exciton and electromagnetic modes in periodic multiple-quantum wells of $CdTe/Cd_{0.87}Zn_{0.13}Te$ with the period equal to either one-half or one-fourth of the light wavelength in the barrier material are experimentally demonstrated by means of reflectivity studies. The measurements are performed for various thicknesses of the cladding layer, incidence angles, magnetic fields, and degree of disorder. Reflectivity spectra as well as energies and radiative lifetimes of the polaritonlike excitations are computed for the relevant structures. It is demonstrated that current theory describes quantitatively the main features of the reflectivity. For multiple-quantum wells with the period equal to one-half of the photon wavelength in the barrier material a strong enhancement of the exciton-photon coupling is observed. This enhancement allows the detection in the frequency domain of the modification of the radiative coupling due to the presence of a dielectric mirror. A considerably different behavior of structures with the period equal to one-half and one-fourth of the light wavelength is predicted to show up in four-wave mixing time-resolved measurements. It is suggested that the description of the luminescence requires a thorough consideration of localization and thermalization dynamics. [S0163-1829(96)00443-2]

I. INTRODUCTION

Recent years have witnessed a growing interest in modification of the coupling between light and electronic degrees of freedom by means of *photon* mode engineering.[1] Semiconductor layered structures, with tailored exciton and electromagnetic excitations, are particularly promising in this context. Indeed, the existence of polariton effects has been put into evidence in $GaAs/Al_xGa_{1-x}As$ and $Ga_{1-x}In_xAs/GaAs$ systems by experimental[2] and theoretical[3] studies of optical response of a single quantum well (QW) placed in a semiconductor microcavity. The microcavities under consideration consisted of a central spacer layer (in which a QW was embedded) and two distributed Bragg reflectors, that is, mirrors in the form of dielectric multilayers, each consisting of two semiconductors with different refraction indexes and thicknesses equal to one quarter of the light wavelength in the corresponding medium.

Apart from a QW in a microcavity, strong polariton effects have been theoretically predicted to occur in long period multiquantum-well (MQW) systems. These are Bragg structures, in which both the real and imaginary part of the refractive index are modulated. In particular, in pioneering works Ivchenko and co-workers[4] examined the interference of light reflected by particular QW's and suggested the existence of a large enhancement of the exciton reflectivity and its radiative recombination rate for structures with a period equal to a half of the light wavelength in the barrier material. Their results have recently been confirmed and extended by Andreani,[5] and in a different formalism by Citrin.[6] In particular, the latter provides a convenient method to determine not only the energies and the radiative lifetimes but also the wave functions of coupled exciton and electromagnetic modes. It is also known that in the case of a single QW, exciton spectra are affected by the thickness of the cladding layer.[7] This is due to the interference of light reflected by the surface and by the QW. Thus it could be expected that the thickness of the cladding layer constitutes an important parameter in a MQW system as well.

In this paper, we present results of reflectivity measurements for long-period MQW's, together with their detailed theoretical description. Our studies were carried out in the spectral region of the ground-state exciton in a long period MQW's of $CdTe/Cd_xZn_{1-x}Te$.[8] Recently, reflectivity studies on similar structures of $CdTe/Cd_xMg_{1-x}Te$ were initiated by Kochereshko *et al.*[9] Our samples consisted of ten thin CdTe wells separated by thick $Cd_{0.87}Zn_{0.13}Te$ barriers. Different types of structures were grown and examined: they had the period and thickness of the cladding layer equal to either one-half or one fourth of the light wavelength in the barrier material. The measurements were performed for various incidence angles, magnetic fields, and degrees of disorder. Our results reveal strong differences in the magnitude, width, and shape of the reflectivity signal in the exciton spectral region depending on the type of the studied structure. By a quantitative calculation of the reflection coefficient, we demonstrate that the rich variety of the observed behaviors can be reproduced accurately by Ivchenko and co-workers theory.[4] Our conclusion is supported by experimental and theoretical studies of disorder effects that might be expected in real samples. In particular, we show how variations of the exciton energy along the various QW's that constitute the

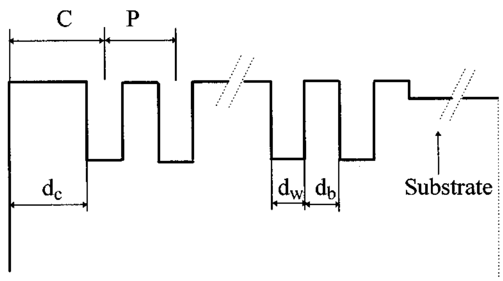

FIG. 1. Schematic illustration of the multiple quantum-well system under consideration. The well width d_w is much smaller than both the barrier d_b and the cladding length d_c. The wells need not to be identical and placed periodically.

structure modify its optical response. In addition, measurements in magnetic fields make it possible not only to enhance the exciton-photon coupling but also to obtain spectra that are not perturbed by the negatively charged exciton X^-.

We also discuss eigenenergies and radiative lifetimes of polariton modes in MQW structures, which we have computed according to the Citrin model.[6] Direct information on these modes not only gives a better insight into the dominant processes involved but also constitutes a step toward understanding photoluminescence and time-resolved measurements.[10]

Our paper is organized as follows. In Sec. II we outline the main features of the theoretical models that will subsequently be applied for the description of experimental results. We discuss also the computed energies and lifetimes of polariton excitations. We then present, in Sec. III, information on growth and characterization of our structures. The experimental findings, together with their theoretical description, are discussed in Sec. IV. The principal conclusions of our work are summarized in Sec. V.

II. THEORETICAL BACKGROUND

A. Reflection and transmission coefficients

We aim at describing the reflection and transmission coefficients, R and T, respectively, in the region of excitonic transitions in a long-period MQW system. In the structures under consideration, shown schematically in Fig. 1, the well thickness d_w is much smaller than the wavelength λ of light in the barrier material, whereas the barrier thickness d_b is of the order of λ. The particular QW's are *not* assumed to be identical.

We start by considering complex amplitudes r and t of reflection and transmission coefficients, respectively, of an isolated QW residing between semi-infinite barriers, $d_b \to \infty$. For photon energy in the vicinity of an exciton resonance we then have, for the TE waves,[4,11]

$$r=\frac{i\Gamma_o}{\hbar\omega_o-\hbar\omega-i(\Gamma_o+\gamma)}, \qquad (1)$$

$$t=1+r. \qquad (2)$$

Here ω_o is the exciton resonance frequency; Γ_o is the energy of the exciton–photon coupling,[11] which is simply related to the radiative lifetime τ_r of an exciton in a single quantum well, $\Gamma_o=\hbar/(2\tau_r)$; and γ describes a nonradiative damping.

In the case of a MQW, interference of waves reflected by particular QW's becomes important. We calculate r_j and t_j, the reflection and transmission amplitudes at frequency ω for a system of j QW's residing in an infinite barrier medium, under assumption that r_{j-1} and t_{j-1} are known. We then obtain general recurrence formulas in the form

$$r_j=r^{(j)}+\frac{(t^{(j)})^2 r_{j-1}\exp(2\,i\varphi^{(j)})}{1-r^{(j)}r_{j-1}\exp(2\,i\varphi^{(j)})}, \qquad (3)$$

$$t_j=\frac{t^{(j)}t_{j-1}\exp(i\varphi^{(j)})}{1-r^{(j)}r_{j-1}\exp(2\,i\varphi^{(j)})}. \qquad (4)$$

In the above formulas $r^{(j)}$ and $t^{(j)}$ are given in Eqs. (1) and (2), respectively, with parameters $\omega_o^{(j)}$ and $\Gamma_o^{(j)}$ corresponding to jth QW, and the phase difference $\varphi^{(j)}=2\pi d^{(j)}/\lambda^{(j)}$, where $d^{(j)}$ is the distance between the centers of jth and $(j-1)$th QW, $\lambda^{(j)}=2\pi c_o/[\omega n_b^{(j)}(\omega)]$, and $n_b^{(j)}(\omega)$ is the refractive index of the intervening barrier. It is assumed here that the background refractive index of the barrier and of the well are either identical, or that their difference is irrelevant since $d_b \gg d_w$.

A particularly appealing prediction of the model in question[4] is a strong increase of the radiative recombination rate in periodic structures ($d_j \equiv P$ for all j) with the period P equal to $\lambda/2$. In such a case the reflection and transmission coefficients r_N and t_N for a set of N QW's are given by Eqs. (1) and (2), respectively, with Γ_o replaced by $N\Gamma_o$.

B. Effect of the cladding layer

The influence of dielectric walls on a radiating dipole has been examined in many contexts, both from classical and quantum points of view.[12] In particular, in the case of an exciton in a single QW, the cladding layer modifies the exciton energy and its lifetime damping according to

$$\hbar\overline{\omega_o}=\hbar\omega_o-r_c\Gamma_o\sin(2\varphi_c), \qquad (5)$$

$$\Gamma_e=\Gamma_o[1-r_c\cos(2\varphi_c)]. \qquad (6)$$

Here $\varphi_c=2\pi C/\lambda_c$, $\lambda_c=2\pi c_o/[\omega n_c(\omega)]$, and

$$r_c=\frac{1-n_c}{1+n_c}, \qquad (7)$$

where $C=d_c+d_w/2$ is the total thickness (see Fig. 1), and $n_c(\omega)$ is the refractive index of the cladding layer.

It is seen that for a typical semiconductor, where $n_c\approx 3$, i.e., $r_c\approx -0.5$, the radiative coupling Γ_e is by a factor 3 greater for a structure with $C=\lambda_c/2$ than in the case of $C=\lambda_c/4$. At the same time, as shown by Zheng *et al.*,[7] the shape of the reflectivity line undergoes a change from emissionlike to absorptionlike.

Similarly strong effects of the cladding layer have been predicted to occur in the case of MQW structures.[4] Taking into account the interference associated with the cladding layer, we arrive at the final expressions for the reflection and transmission coefficients of the structure with N QW's,

$$R=\left|\frac{r_c+r_N\exp(2\,i\varphi_c)}{1+r_c r_N\exp(2\,i\varphi_c)}\right|^2, \tag{8}$$

$$T=n_c\left|\frac{t_c t_N\exp(\,i\varphi_c)}{1+r_c r_N\exp(2\,i\varphi_c)}\right|^2, \tag{9}$$

where

$$t_c=1+r_c. \tag{10}$$

In the particular case of a structure with the period equal to $\lambda/2$, r_N is given by Eq. (1) with Γ_o replaced by

$$\Gamma_e=N\Gamma_o[1-r_c\cos(2\varphi_c)], \tag{11}$$

where $\cos(2\varphi_c)$ describes the influence of the cladding layer; $\cos(2\varphi_c)=1$ for $C=n\lambda_c/2$ and $\cos(2\varphi_c)=-1$ for $C=(2n+1)\lambda_c/4$ with n being an integer.

C. Polariton modes

For the description of luminescence or time-resolved experiments, such as radiative lifetime or four-wave mixing, it is convenient to determine directly energies and damping of the coupled exciton-photon excitations. In large-period MQW's, where carrier tunneling may be neglected, the transfer of excitons between QW's takes place through two mechanisms: (i) the Förster mechanism, which operates for excitons with in plane wave vector $k_\parallel$ greater than $k_p=2\pi/\lambda$, the wave vector of light in the barrier material. This interaction is driven by the instantaneous dipole-dipole coupling, and for free excitons decays exponentially with the QW separation. (ii) A polaritonlike mechanism for excitons with $k_\parallel<k_p$, for which the coupling proceeds *via* the coherent emission and reabsorption of a photon. This is a long-range interaction which oscillates with the QW separation. The Förster mechanism may become a dominant process of exciton transfer between *nonequivalent* QW's, i.e., with different exciton energies. Here we consider periodic structures, made of nominally identical QW's, and therefore only the polaritonlike mechanism of the exciton transfer is taken into account.

For a single quantum well imbedded between two semi-infinite barriers, following Citrin[6] and Andreani,[5] we define the radiative coupling of the $1se_1h_1$ exciton to the radiation field with the in-plane polarization vector along $\mathbf{k}_\parallel$ as $\Gamma_o^{(L)}(k_z)=\Gamma_o k_z/k_\parallel$ and perpendicular to $\mathbf{k}_\parallel$ as $\Gamma_o^{(T)}(k_z)=\Gamma_o k_p/k_z$, where $k_z=\sqrt{k_p^2-k_\parallel^2}$ and

$$\Gamma_o=\pi e^2 f_{xy}/(m_o c_o n_b), \tag{12}$$

with f_{xy} being the oscillator strength.

Now we consider a set of N identical QW's with equal thickness d_w, and distances between their centers (period) P, where $P\gg d_w$. The structure is embedded in an infinite medium so that no influence of the cladding layer is taken into account. We neglect disorder effects as well as carrier tunneling or hopping over the barriers. Due to the radiative energy transfer, an exciton with a given $k_\parallel$ and polarization s is delocalized over all QW's. There are N such collective excitations with eigenvalues

$$E_i^{(s)}(k_\parallel)=E_{\rm ex}(k_\parallel)+\epsilon_i^{(s)}(k_\parallel), \tag{13}$$

and radiative recombination rates $\Gamma_i^{(s)}(k_\parallel)$, where $i=1,2,\ldots,N$; $s=L$ or T, and $E_{\rm ex}(k_\parallel)$ is the exciton energy for a single QW. If $\Gamma_i^{(s)}(k_\parallel)$, and the relative occupancy of the states $E_i^{(s)}(k_\parallel)$ are known then the luminescence intensity in the direction corresponding to $k_\parallel$ can be calculated. This occupancy is given by the Boltzmann distribution, provided that full thermalization over the polariton modes is achieved.

Following Citrin[6] it is easy to show that ϵ_i and Γ_i are solutions of the equation $\det[A(\epsilon,\Gamma)]=0$, where A is a $N\times N$ matrix. In the space representation (an exciton localized in one well), and in the exciton-pole approximation, which is expected[5] to be accurate for the values of Γ_o and N of interest here, the matrix A assumes the form

$$A_{\ell\ell'}=a_{\ell\ell'}-[\epsilon-\,i(\Gamma+\gamma)]\delta_{\ell\ell'}, \tag{14}$$

with

$$a_{\ell\ell'}=-\,i[\Gamma_o^{(s)}(k_z)\exp(\,ik_z|\ell-\ell'|P)], \tag{15}$$

where $\ell,\ell'=0,1,\ldots,N-1$. Note that A is symmetric but not Hermitian. Note also that ϵ and Γ are determined totally by the product of k_z and P. Accordingly, the model gives complex eigenvalues as a function of both P/λ (for a given $k_\parallel$) and $k_\parallel$ (for a given P/λ). In an equivalent approach by Andreani,[5] the mode energies and lifetimes are given by poles of the transmission amplitude. However, an important aspect of the Citrin model discussed here is that it gives additionally information on the eigenstates, i.e., on the probability of finding the exciton in a given QW.

We now consider the simple case of a double quantum well, $N=2$. Then the two modes in questions are the symmetric and antisymmetric linear combinations of the states obtained by localizing the exciton in one or in the other QW. Their energies for $\gamma=0$, shown also in Fig. 2, are[6]

$$E_\pm=E_{\rm ex}(k_\parallel)+\Gamma_o^{(s)}(k_z)[\pm\sin k_zP-\,i(1\pm\cos k_zP)]. \tag{16}$$

It is seen that the radiative width that is measured in transmission or reflectivity measurements at normal incidence becomes an oscillatory function of the distance between the two QW's. Hence for $k_zP=2\pi$ modulo 2π the radiative linewidth of the symmetric mode is twice the one for a single QW, whereas the coupling to the antisymmetric mode vanishes. In contrast, for $k_zP=\pi$ modulo 2π it is the antisymmetric mode which is coupled to the electromagnetic field. In both cases one retrieves the result of Ref. 4, i.e., that for $P=\lambda/2$ modulo $\lambda/2$ the radiative width is enlarged by a factor equal to the number of QW's. It is worth noting that the shift of the two eigenmodes shows the same oscillatory behavior with the QW separation but in quadrature. Thus the splitting between the symmetric and antisymmetric modes reaches its maximum value of $2\Gamma_o$ for $k_zP=\pi/2$ modulo π and becomes zero for $k_zP=\pi$ modulo π.

The computed radiative couplings and energy shifts of the eigenmodes for a set of ten QW's are shown in Fig. 3 for

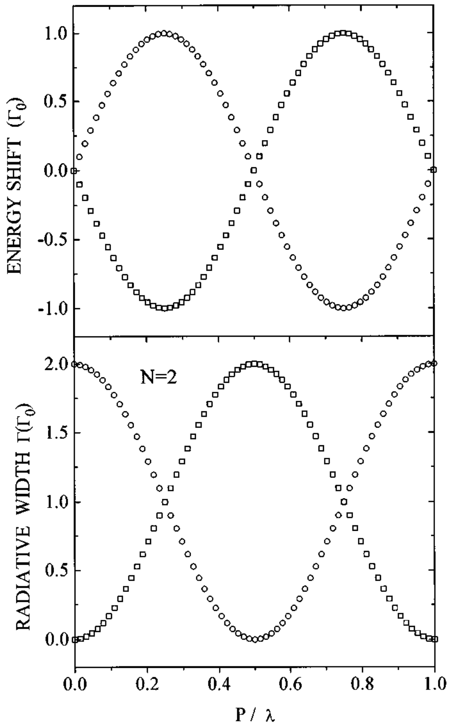

FIG. 2. Calculated energy shifts (upper panel) and radiative widths (lower panel) for polaritons with zero in-plane momentum for two identical QW's as function of the ratio of the period P to the wavelength λ: circle (square) symmetric (antisymmetric) eigenstates.

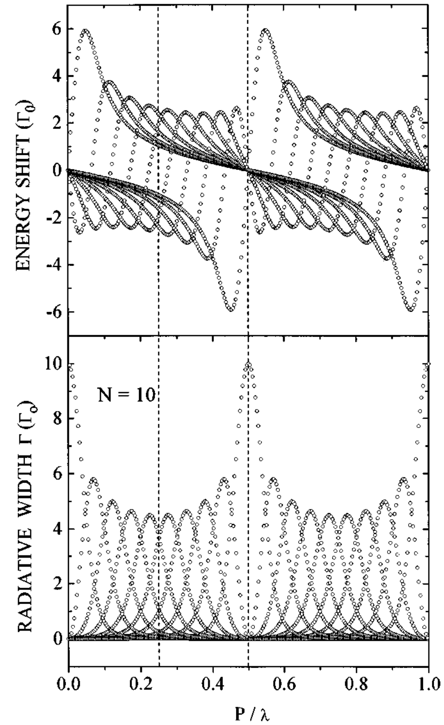

FIG. 3. Calculated energy shifts (upper panel) and radiative widths (lower panel) for polaritons with zero in-plane momentum for a stack of ten identical QW's as function of the ratio of the period P to the wavelength λ.

$k_{\parallel}=0$ as a functions of the period P expressed in wavelength unit λ. These results agree with those calculated by Andreani[5] for $P/\lambda=1/8\pi$ by using a transfer-matrix approach. We also retrieve the result of Ref. 4 for $P=\lambda/2$ modulo $\lambda/2$: there is only one mode which couples to the radiation field, and the coupling is enhanced by a factor of $N=10$. For $P=\lambda$ it is the zone-center mode, with the in-phase excitations of all QW's, which is coupled to the photons. For $P=\lambda/2$, in turn, the coupling exists for the zone-boundary mode with a π phase shift at adjacent QW's. We make the same remark as for the double QW: for the period $P=\lambda/2$ modulo $\lambda/2$ the energy shift of all the modes vanish. However, away from this point at $P/\lambda\approx 0.43$ the energy shift can be as large as $6\Gamma_o\approx 7$ K for CdTe/$Cd_xZn_{1-x}Te$. These general properties of the modes should not be significantly affected by the presence of a finite cladding layer.

The modes in question should be directly observed in, for instance, luminescence under resonant excitations, Brillouin or Rayleigh scattering, and time-resolved measurements. In particular, a rather dramatic increase of the radiative decay rate in samples with $P=\lambda/2$, and the presence of beating effects for other periods are expected to be visible in time-resolved four-wave mixing experiments. For a particular case of the $\lambda/4$ structure, there are two coupled modes that contain 40% of the total oscillator strength, $10\Gamma_o$. Their splitting, that is, the beating frequency—is $3.9\Gamma_o$, which corresponds to about 0.4 and 0.1 meV in the case of CdTe/$Cd_xZn_{1-x}Te$ and GaAs/$Al_xGa_{1-x}As$ MQW's, respectively. For nonresonant excitation, and provided that localization effects can be disregarded, the results displayed in Fig. 3 suggest a strong and complex dependence of the luminescence intensity on the emission angle and on the value of P/λ.[10] For example, the luminescence intensity at sufficiently low temperatures and in the direction normal to the surface is predicted to undergo a marked maximum for $P/\lambda=0.43$. This is because for this value of P/λ, the thermalized excitons should occupy a rather narrow region of the phase space around $k_{\parallel}=0$.

D. Absorption spectra and exciton oscillator strength

Due to the coherence effects discussed above (Secs. II A and II B) the reflectivity spectra of MQW's depend strongly on both the period and the cladding layer thickness. Thus in order to determine the oscillator strength, a careful analysis of the reflectivity spectra is needed. The transmission spectra are frequently used to determine exciton oscillator strength. The latter is assumed proportional to the integrated absorbance line

$$A=\int \ln\left(\frac{T_{\max}}{T}\right) d\hbar\omega, \tag{17}$$

where the integration is carried over on the exciton line, and $T_{\max}$ is the background transmission coefficient taking into account the reflection at sample surfaces. We will now discuss the validity of this method. In the case of a single QW, or of a Bragg structure of N QW's with $P=\lambda/2$ modulo $\lambda/2$ (which behave as a single QW with radiative coupling $N\Gamma_o$), the transmission coefficient is calculated using Eqs. (1), (2) and (9). It is easy to show that the area $A=2\pi\Gamma_e$ is independent of the nonradiative coupling coefficient γ, Γ_e is

TABLE I. Nominal and measured by x-ray-diffraction length of the period P and the thickness of the cladding layer C of the studied samples with ten wells. The wavelength λ of the light in the barrier material of $Cd_{0.87}Zn_{0.13}Te$ at the energy of the exciton in the quantum well of 10-nm CdTe is evaluated to be 260 nm.

Sample	Nominal cladding C	Nominal period P	Period P (nm)
I	λ/2	λ/2	128
II	λ/2	λ/4	67.5
III	λ/4	λ/2	133.7
IV	λ/4	λ/2	123.2
V	λ/2	λ/2	135.9
VI	λ/2	λ/4	61.6

the radiative coupling coefficient given by Eq. (11). Thus it changes by a factor $(1+r_c)/(1-r_c)$ close to 3 when the cladding layer thickness varies from $\lambda/4$ to $\lambda/2$. For a period different from $\lambda/2$ modulo $\lambda/2$, the effects of the sample surface on the radiative coupling of the different QW's tend to cancel each other and various simulations show that, if the number of QWs is large enough, $N>4$, the area A is close to $2\pi N\Gamma_o$.

III. SAMPLE GROWTH AND CHARACTERIZATION

A series of ten-period MQW's with various barrier and cladding layer thicknesses were grown by molecular-beam epitaxy onto (100)-oriented $Cd_{1-y}Zn_yTe$ substrates with $y=12\%$. The value of y was checked by x-ray diffraction and the optically measured band gap. In order to assure a pseudomorphic growth of the MQW's, the layer widths, d_w and d_b, as well as the Zn concentration in the barriers, x, was designed according to the following principles. First, all individual layers were grown with the thickness below its critical value t_c for the formation of misfit dislocations. As shown previously[13] for the CdTe/ZnTe system with the lattice mismatch of 6.4%, and t_c is 40 nm and 650 nm for CdTe on $Cd_{0.88}Zn_{0.12}Te$ and $Cd_{0.87}Zn_{0.13}Te$ on $Cd_{0.88}Zn_{0.12}Te$, respectively. Second, the Zn concentration in the barrier was chosen to be $x=0.13$, as such a value results in a strain symmetrized structure, in which the average strain is close to zero, so that the condition of pseudomorphic growth is fulfilled for arbitrary thickness of the MQW structure.

The above procedure has made it possible to avoid strain relaxation in our thick structures, despite a relatively large lattice mismatch between the barriers and the wells, $\Delta a/a\approx 0.7\%$. Additionally, in order to improve the interface quality and to make reflection high-energy electron-diffraction (RHEED) oscillations visible during the whole process of epitaxy, a stop and grow method has been employed.

The thickness of QW's is $d_w=10$ nm, resulting in an e_1h_1 exciton energy $\hbar\omega_o=1607$ meV. Assuming the same background dielectric constant for the barrier alloy and the QW's $\epsilon_b=n_b^2=8.8$, we have $\lambda/2=130$ nm.

Parameters that characterize the studied samples are presented in Table I. The mean periods have been determined by x-ray diffraction. Note that our definitions of the period length $P=d_b+d_w$ and the thickness of the cladding layer

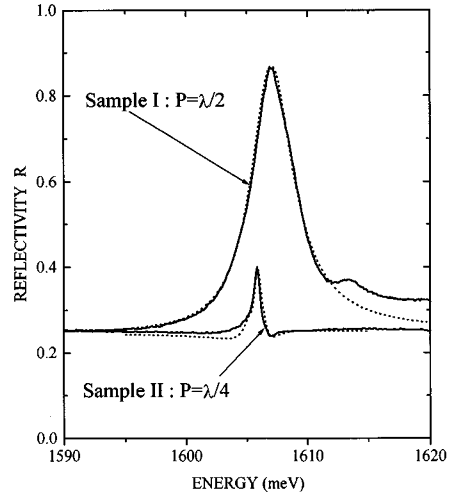

FIG. 4. A comparison of the reflectivity spectra in the region of the 1s e_1h_1 exciton for two stacks of ten QW's of CdTe/$Cd_xZn_{1-x}Te$ with the same thickness of the cladding layer $C=\lambda/2$ but with the periods P close either to $\lambda/2$ (sample I) or to $\lambda/4$ (sample II) in a magnetic field of 5 T for σ^- light polarization. The dashed lines are theoretical, $\Gamma_o=0.12$ meV, and $\gamma=0.3$ meV.

$C=d_c+d_w/2$ differ from those used by Ivchenko and co-workers.[4]

IV. EXPERIMENTAL RESULTS AND DISCUSSION

A. Experimental method

Reflectivity spectra were measured for various incidence angles by standard techniques with the use of an iodine lamp and an optical cryostat. The back surface of the substrate was not polished, but prior to measurements it was etched in HCl in order to remove a gallium layer that remained from the growing process. The samples were immersed in a bath of liquid helium pumped down to 1.7 K. Corrections for the spectral response of the apparatus were determined by measuring transmission of a reference substrate and reflectivity by a gold mirror. Absolute calibration of the reflectivity signal was sometimes unreliable. In such cases, the magnitude of the signal was corrected by a multiplicative factor to give $R=0.25$ at energies above the band gap of the barrier material.

B. Bragg vs anti-Bragg structures

Figure 4 shows experimental and calculated reflectivity spectra for sample I and II with period P equal to 0.50λ and 0.26λ, i.e., close to $\lambda/2$ and $\lambda/4$, respectively, and a cladding thickness $C=0.50\lambda$, where λ is the wavelength of light in the barrier material. Both samples contain $N=10$ QW's, and the measurements were taken for normal incidence. The two spectra are seen to be strikingly different. In particular, the exciton resonance is much stronger and wider for sample I (λ/2;λ/2) in the notation $(C;P)$, than in the case of sample II with (λ/2;λ/4). Generally speaking, this different behavior results from the fact that for samples, in which the Bragg condition is met, $P=\lambda/2$, interference of light reflected by N QW's is constructive. By contrast, for $P=\lambda/4$ the inter-

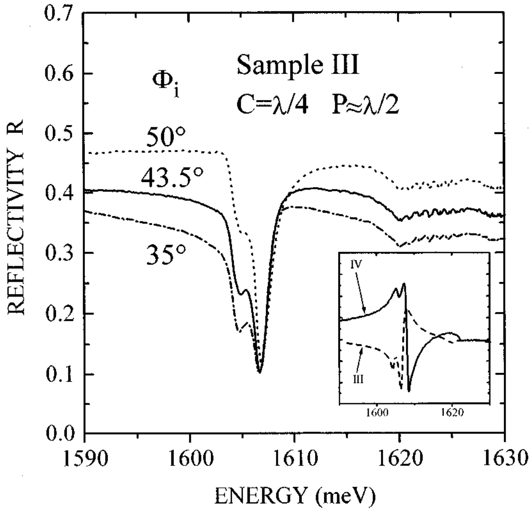

FIG. 5. Reflectivity spectra in the region of the 1*s* e_1h_1 exciton at various incidence angles ϕ_i for a stack of ten QW's of CdTe/$Cd_xZn_{1-x}Te$ with the thickness of the cladding layer λ/4 and the period *P* slightly greater than λ/2 (sample III). The light is polarized perpendicularly to the plane of incidence. Inset: reflectivity at normal incidence for III (dashed line) and sample IV (full line) with period (134 nm), slightly greater (III) or (123 nm), slightly less (IV) than λ/2. For both samples the cladding thickness is λ/4.

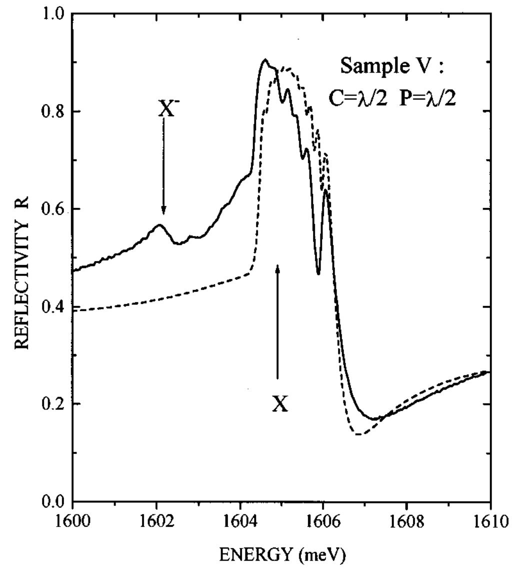

FIG. 6. Reflectivity in the region of the $1se_1h_1$ exciton for a stack of ten QW's of CdTe/$Cd_xZn_{1-x}Te$ with the thickness of the cladding layer and the period close to λ/2 (sample V). The multipeak structure is assigned to gradual changes of the exciton energy along the MQW structure. The dashed line is calculated under the assumption that the QW energies increase uniformly from 1604.6 (close to the surface) to 1606.1 meV; meanwhile their separations decrease from 140.4 nm to 131.4 nm in agreement with x-ray and RHEED data; Γ_o=0.1 meV and γ=0.05 meV for all QW's. In order to fit the background reflectivity an additional reflection (15%) from the back surface of the substrate was taken into account.

ference effect is destructive. However, a deeper insight is provided by analysis of the coupled exciton-photon modes, as discussed in Sec. II C.

In order to compare experimental results and theoretical expectations, the reflection coefficients were calculated numerically from Eqs. (3) and (8). Since only a narrow frequency range around the exciton resonance is of interest here, the frequency dependence of λ has been neglected. As shown by dashed lines in Fig. 4. a remarkably good agreement between the experimental findings and the calculated curves has been obtained by taking the radiative and nonradiative damping coefficients $\Gamma_o = 0.12$ meV and $\gamma = 0.3$ meV, respectively, for both samples. In the case of (λ/2;λ/4) (sample II) the half width at half maximum (HWHM) is only 0.35 meV, and comparable to that observed in best quality single QW's. It is given as a good approximation by the sum of Γ_o and γ. On the opposite for sample I the HWHM is as large as 2.1 meV, in agreement with Eqs. (1) and (11) for $N = 10$, $\cos(2\varphi_c) = 1$, and $r_c = -0.5$.

The spectra shown in Fig. 4 were taken in the magnetic field of 5 T with left circularly polarized light. They differ from those obtained at zero field[8] by the absence of an additional line (2.7 meV below the free-exciton one) due to the formation of charged exciton X^-.[14] Because of residual doping in the barrier this line is always present in large-period CdTe/$Cd_xZn_{1-x}Te$ MQW's. Its suppression by application of a magnetic field[14] allows a better analysis of the free-exciton line shape. The slight increase of the radiative coupling coefficient Γ_o from its 0.1 meV value at zero field[8] to 0.12 meV at 5 T could be due to the magnetically induced shrinking of the 1*s* exciton wave function. These values of Γ_o are consistent with those resulting from the measured and evaluated oscillator strengths of the ground-state exciton transition in QW's of CdTe.[15] This supports even further our conclusion about general accord between experimental results and theoretical model. On the other hand, theoretical analysis of the obtained values of γ is more difficult, as it may contain a contribution from extrinsic effects such as inhomogeneous broadening of exciton energies in our MQW system. We shall come back to this point in Sec. IV D.

C. Influence of the cladding layer

As we mentioned in Sec. II B, the shape of the reflectivity spectra varies periodically with the cladding thickness *C*. For $C = n\lambda/2$ (*n* integer) the light reflected by the quantum wells and by the surface are in phase, and the reflectivity resonance is emissionlike. For $C = (2n+1)\lambda/4$, however, the reflectivity resonance is expected to be absorptionlike, since the two reflected waves have opposite phases. The latter is illustrated in Fig. 5. The inset shows the reflectivity spectra of samples III and IV, both having a thickness of the cladding layer equal to λ/4 while *P* close to λ/2. The MQW periods are equal to 134 and 123 nm, i.e., *P* is slightly larger and smaller than λ/2, for samples III and IV, respectively. It is seen that for this value of *C*, $C = \lambda/4$, the shape of the reflectivity spectrum depends rather dramatically on the exact value of the period.

For a sample with a period greater than λ/2, the Bragg resonance condition can be met by measuring reflectivity under oblique incidence. In such a case the Bragg condition becomes $P\cos\phi_B = \lambda/2$, where ϕ_B is the angle between the light direction inside the sample and the surface normal. Fig-

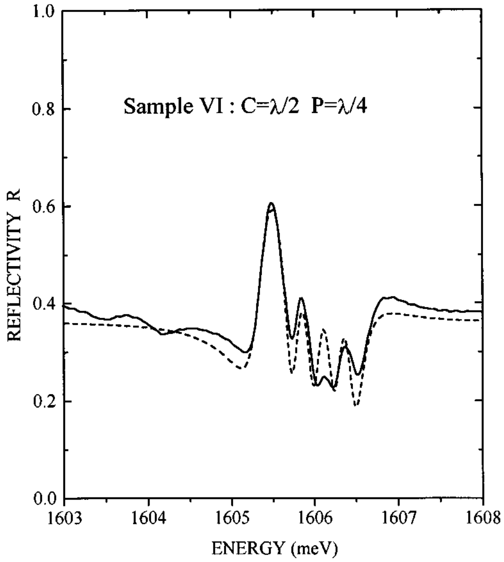

FIG. 7. Reflectivity in the region of the $1se_1h_1$ exciton for a stack of ten QW's of CdTe/Cd_xZn_{1-x}Te with the thickness of the cladding layer λ/2 and the period close to λ/4 (sample VI). The multipeak structure is assigned to gradual changes of the exciton energy along the MQW structure. The dashed line is calculated under the assumption that the exciton energy in the QW's increases uniformly from 1605.4 meV (close to the surface) to 1606.4 meV, while the QW separation is constant and equal to the period $P=61.6$ nm, as measured by x-ray diffraction; $\Gamma_o=\gamma=0.1$ meV.

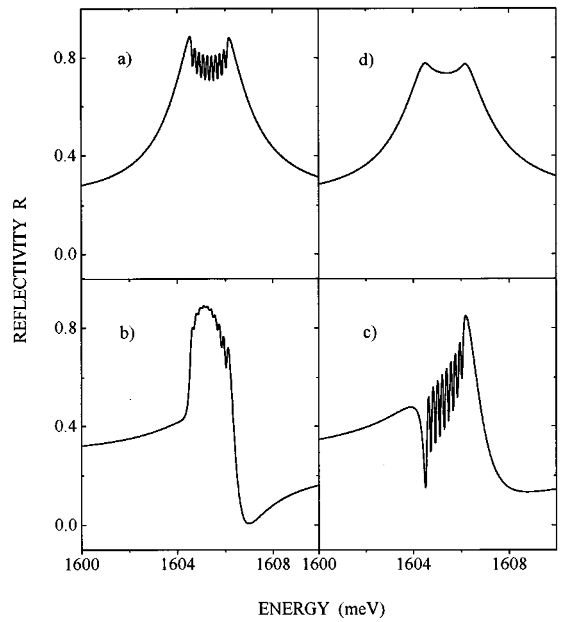

FIG. 8. Calculated reflectivity in the region of the $1se_1h_1$ exciton for a stack of ten QW's of CdTe/Cd_xZn_{1-x}Te with the thickness of the cladding layer $C=\lambda/2$ and the average period P close to λ/2. (a) $C=P=\lambda/2$; the nonradiative damping $\gamma=0.05$ meV; the exciton energies in the QW's either increase or decrease uniformly from 1604.6 (surface side) to 1606.1 meV. (b) The exciton energies in the QW's increase uniformly from 1604.6 to 1606.1 meV; meanwhile, the QW separation decreases from 140.4 to 131.4 nm. (c) Same as (b) but the in reverse order; both the QW thicknesses and their separation decrease from the surface side toward the substrate. (d) Same as (a), but $\gamma=0.2$ meV.

ure 5 shows the reflectivity spectra of sample III for three slightly different incident angles ϕ_i. The light polarization is perpendicular to the plane of incidence. The spectrum measured for $\phi_i=43.5°$ is very symmetric (no overshoot in the wings) which, according to the numerical simulations, shows that the Bragg resonance condition is nearly exactly fulfilled. Since for $\phi_i=43.5°$, $\phi_B=13.6°$, and $\cos\phi_B=0.972$, we see that the effective period $P\cos\phi_B$ has been reduced by only 2.8% in respect to its value at normal incidence. Moreover, from the angle corresponding to the Bragg resonance a precise value of the refractive index and thus of the dielectric constant of the barrier alloy at the exciton wavelength can be determined. The value obtained in this way $\epsilon_b=n_b^2=8.8\pm0.2$ has been adopted for all our calculations. We note that this value is somewhat smaller than the dielectric constants of CdTe and ZnTe, 9.65 and 9.67, respectively, given by Segall and Marple.[16] It is, however, in an agreement with the dielectric constant $\epsilon=9$ estimated by Segall[17] for CdTe at the exciton energy of 1.6 eV.

It worth noting that the thickness of the cladding layer not only affects the shape of the line but also its broadening. For sample III, under the resonant Bragg condition, HWHM Γ_e is given by Eq. (11) with $\cos(2\varphi_c)=-1$ and $r_c=-0.61$ for the incidence angle of 43.5°. These numbers imply that the radiative linewidth should be three times smaller in structures with a cladding $C=\lambda/4$ than in MQW's, in which both the cladding and the period are λ/2. The data of Fig. 5 (sample λ/4;λ/2) illustrate this strong reduction of the radiative coupling. The measured linewidth (HWHM) is 1.1 meV, i.e., two times smaller than for sample I (λ/2;λ/2) and in good agreement with the prediction of Eq. (11) when taking $\gamma=0.44$ meV, a value similar to those determined for the other samples.

D. Disorder effects

Finally, we turn to the important question of the sensitivity of the studied effects to disorder that may be present in real structures. We specify to vertical disorder, i.e., we consider an eventual distribution of resonance energies for the excitons in the different QW's. Figure 6 presents a complex multipeak spectrum that has been recorded for a sample with a nominal period and cladding designed to be λ/2 (sample V). Similar data for sample VI with the nominal period λ/4 is shown in Fig. 7. We know from the RHEED data that the growth rate of these structures slowly and monotonously changes during the epitaxy process, so that in sample V the first QW was by about 1.5 ML (0.45 nm) narrower than the last one and similarly the QW separation increased by about 7% along the structure. In the simulation shown in Fig. 6 the average period $\overline{P}=136$ nm was taken from the x-ray measurements. Furthermore, the energies of QW's were chosen to vary linearly from 1604.6 meV at the sample surface to 1606.1 meV, while their separation to decrease from 140.4 nm to 131.4 nm, in agreement with the RHEED data. A good fit to the experiment is obtained assuming the same nonradiative damping coefficient γ for the ten QW's. A similar fit is shown in Fig. 7 for sample VI, a structure with the period close to $\lambda/4$.

In order to quantify the effect of disorder we performed a series of simulations, representative results of which are summarized in Fig. 8. It is seen that when the period is not exactly matched to $\lambda/2$ the spectra vary strongly with the sign of the gradient of the QW exciton energy along the structure. As could be expected, and Fig. 8(d) shows, for appropriately large values of the nonradiative damping coefficient γ the fine structure tends to disappear. However, the resulting line shape is by no means Lorentzian. This indicates that in our good quality structures, such as sample I, for which a truly Lorentzian line shape is observed (see Fig. 4), the dispersion in the widths of QW's is negligibly small.

V. CONCLUSIONS AND OUTLOOK

Our results have provided experimental verification of several aspects of current theory of polariton properties in long period multiquantum well structures. In particular, we have put into evidence a strong enhancement of the exciton-photon coupling for MQW's with a period equal to a half of the photon wavelength. Then, as predicted by Ivchenko,[4] the set of N QW's behave as a single oscillator with the radiative coupling N times greater than for a single QW, and the exciton reflectivity spectrum shows a broad Lorentzian line. This enables the observation in the frequency domain of an effect[11] which is usually searched for in the time domain, namely, the modification of the radiative coupling induced by a dielectric mirror. This modification is associated with the surface of the structure. In particular, when thickness of the cladding layer is varied from $\lambda/2$ to $\lambda/4$, the radiative coupling decreases by a factor of 3, which results in a large diminution of the exciton linewidth. We also showed that a precise determination of the exciton oscillator strength needs a careful analysis of the reflectivity and transmission spectra, taking into account the QW separations and the cladding layer thickness. Furthermore, our results show how randomness of real structures affects the studied phenomena, and that a rather large vertical disorder is necessary to quench the formation of a superradiant state in the $P=\lambda/2$ structures.

Additionally, in order to visualize the polariton modes, their energies and radiative lifetimes have been computed as a function of the MQW period P and the wave vector $k_{\parallel}$. These results constitute a good starting point for a description of a number of experiments such as luminescence, Brillouin or Rayleigh scattering, and time-resolved measurements. In particular, a rather dramatic increase of the radiative decay rate in samples with $P=\lambda/2$ and the presence of beating effects for other periods is expected to be visible in time-resolved four-wave mixing spectroscopy. While a first attempt to detect these effects has been only partly successful,[18] more recent four-wave mixing measurements on a MQW system of $GaAs/Al_xGa_{1-x}As$[19] started to reveal the presence of the polariton modes discussed here.

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