Resonant Bragg reflection of light from ZnTe-based structures with embedded CdTe monolayers

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Abstract. An electrodynamic model describing Bragg reflection of light from semiconductor structures with ultrathin planar layers embedded in bulk material matrix is developed. The layers are assumed to exhibit resonant optical properties in a spectral range near the short-wavelength bandgap edge of bulk material. Model calculations of the reflection spectra are performed at typical parameter values of the resonant Bragg structure containing monolayers of CdTe in bulk ZnTe.

1. Introduction

It became clear that electronic and optoelectronic devices with an active region containing planar layers in the form of extremely narrow quantum wells localizing excitons have significant advantages over similar devices with relatively wide wells [1]. Of particular interest [2] are in this case structures formed by wide-gap II–VI compounds where the binding energy of excitons is comparable or even exceeds thermal energy at room temperature. In such systems, one can expect the manifestation of exciton properties in the temperature range interesting for applications. To enhance the interaction of excitons with light, it was proposed in [3] to use a system of equidistant quantum wells in which the optical thickness between adjacent wells is comparable to the wavelength of the exciton resonance. Studies of the most recent years convincingly show that ultra-thin planar layers with a thickness of only a few monolayers (up to single monolayer) embedded in bulk material and possessing dipole-active optical excitations are also very promising for new ways to control the light-matter interaction [4-6].

In this paper, the object under study is a structure consisting of CdTe monolayers embedded in ZnTe bulk matrix. The main attention is paid to the development of an approach to calculating the resonance reflection spectra for such a specific situation when it is problematic to discuss the formation of quantized exciton states in the spirit of a theoretical approach [3]. Focusing on the available preliminary experimental results [2], we assume that an exciton excited in bulk material (ZnTe) can be bound on the ultrathin layer of the implanted material (CdTe) which plays the role of an extended flat defect. In this case, it would be reasonably to assume that the region of spatial localization of the exciton in directions perpendicular to the layer should not exceed, in order of magnitude, the Bohr radius of the bulk exciton, i.e. should be significantly less than the characteristic wavelength of light in the bulk material. Then, from the point of view of a purely optical problem, one can consider the structure as consisting of singular layers in which resonant dipole polarization can be excited.
In this work, model calculations of the resonant reflection spectra of light for a structure containing both a single singular layer and a system of several ordered layers are performed. The calculations took into account effects due to bulk exciton states near the fundamental optical absorption edge of the base material. Attention is paid to substantial enhancement of the resonant reflection signal when using a Bragg structure, which is in qualitative agreement with similar conclusions [3] as applied to systems with standard quantum wells.

2. Problem statement
Let us consider a planar structure whose interface planes are perpendicular to \(-\vec{z}\)-direction in the coordinate system given by the \((\vec{e}_x, \vec{e}_y, \vec{e}_z)\) orts. Taking into account the spatial homogeneity of the structure along the \(x, y\) directions we can write the Maxwell’s equations containing \(\nabla \times\) operators as follows:

\[
(i k_\tau + e_z \nabla_z) \times E(z) = i k_0 B(z); \tag{1}
\]
\[
(i k_\tau + e_z \nabla_z) \times B(z) = -i k_0 \varepsilon_b E(z) + \frac{4\pi}{c} J(z); \tag{2}
\]

where \(E\) and \(B\) are the electric field and magnetic induction vectors, respectively, \(J\) is the polarization current density vector, \(k_\tau\) means the tangential component of the incident light wave vector, \(\varepsilon_b\) is the background permittivity of the medium, where the polarization current generates, \(c\) is the speed of light in vacuum, \(k_0 \equiv \omega/c\) is the wave number at the frequency \(\omega\). The vector \(B(z)\) \((F = E, B, J)\) in (1,2) represents amplitudes \(F(z) \equiv F(\omega, k_\tau; z)\) of monochromatic fields in the form \(F(r; \tau) = \text{e}^{i k(\vec{r}; -\tau)}\), \(\vec{r}_\tau = x \vec{e}_x + y \vec{e}_y\).

We’ll assume that the region of spatial localization (along \(z\) axis) of the polarization current generated in the vicinity of a certain point \(z = \zeta\) is significantly less than the characteristic wavelength of light in the medium. Then from a formal point of view, we can speak of a singular behavior of \(J(z)\) at this point. Without specifying a mechanism of the singular current density emergence within the framework of a certain microscopic model, we will consider \(J(z)\) as a generalized function in the form of expansion it in a series containing \(\delta\)-functions \((\delta \equiv \delta^{(0)}\) and their derivatives \(\delta^{(n)}\) \((n = 1, 2, ... \infty)\):

\[
J(z) = \sum_{n=0}^{\infty} j^{(n)}(z - \zeta) \tag{3}
\]

In such a case, the solution of equations (1, 2) for amplitudes \(F(z)\) should be sought in a corresponding class of generalized functions with zero support [7]

\[
F(z) = F^R(z) + \sum_{n=0}^{\infty} f^{(n)}(z - \zeta), \tag{4}
\]

where amplitudes \(F^R\) refer to regular (non-singular) contributions of fields. The coefficients \(j^{(n)}\) in expression (3) should be given by specific constitutive equations which linearly include both regular \(E^R(\zeta \pm 0)\) and singular \(e^{(n)}\) contributions of the electric field strength \(E(z)\). The problem in a similar formulation was considered earlier in [8] as applied to the analysis of corrections to the Fresnel light reflection coefficients regarding a thin (singular) near-surface transition layer. At the same time, phenomenological parameters of the transition layer were introduced into constitutive equations, the choice of which made it possible to adequately describe a wide variety of situations discussed in the framework of rigorous microscopic approaches.

As a result of simple transformations (by vector and scalar multiplying equations (1, 2) by the \(e_z\) ort) and integrating the resulting equations along \(z\) coordinate over an infinitesimally thin layer of the thickness \(l \to 0\) we have the boundary conditions (BC) in the form

\[
E_\tau(\zeta + 0) - E_\tau(\zeta - 0) = -i k_0 e_z \times b_{\tau}^{(0)} - \frac{i k_\tau}{k_0 e_B} [k_\tau \times b_{\tau}^{(0)}]_z + \frac{4\pi}{k_0 e_B} k_\tau j_\tau^{(0)} \tag{5}
\]
\[
B_\tau(\zeta + 0) - B_\tau(\zeta - 0) = i k_0 e_B e_z \times e_\tau^{(0)} + \frac{i k_\tau}{k_0} [k_\tau \times e_\tau^{(0)}]_z - \frac{4\pi}{c} e_z \times j_\tau^{(0)} \tag{6}
\]
where the subscript $\tau$ means a vector tangential component on the interface plane.

Moreover, additional relations between the coefficients of singularities follow from independence of the contributions to Eqs. (1,2) of the terms containing the $\delta$-function derivatives of different order:

$$e^{(n)}_\tau = -ik_0 e_z \times b^{(n+1)}_\tau - \frac{ik_\tau}{k_0} \left[ k_x \times b^{(n+1)}_\tau \right] + \frac{4\pi}{k_0c} k_{\phi} f^{(n+1)}_\tau$$  \hspace{1cm} (7)

$$b^{(n)}_\tau = i k_0 \varepsilon_0 e_z \times e^{(n+1)}_\tau + \frac{ik_\tau}{k_0} \left[ k_x \times e^{(n+1)}_\tau \right] + \frac{4\pi}{c} e_z \times j^{(n+1)}_\tau$$  \hspace{1cm} (8)

Specific expressions for the reflection coefficients can be obtained if we restrict ourselves to a certain finite number of singular terms in formulae (5) to (8). The choice of such terms should correspond to a certain parameterization of the singular layer properties, which can be specified only within the framework of a specific microscopic model. However, in those cases when there are difficulties in building a sufficiently rigorous microscopic theory that meets specifics of technological conditions for preparing samples, our proposed approach seems to be a significant initial step in developing a theoretical description of the experimental data on reflection of light from structures containing monolayers.

To highlight the fundamental points in the reflection spectra formation for a system of periodically located singular layers, we restrict ourselves to the simplest situation when the singularities in the tangential field components, $E_\tau$ and $B_\tau$, are ignored, so that the current density (3) contains no terms with the $\delta$-derivatives. Then BC (5, 6) are reduced to the form

$$E_\tau (\zeta + 0) - E_\tau (\zeta - 0) = \frac{4\pi}{k_0\varepsilon_0 c} k_{\phi} f^{(0)}_\tau$$  \hspace{1cm} (9)

$$B_\tau (\zeta + 0) - B_\tau (\zeta - 0) = -\frac{4\pi}{c} e_z \times j^{(0)}_\tau$$  \hspace{1cm} (10)

From equations (9, 10) we see that the presence of a singular polarization current $j^{(0)}_\tau$ leads to a violation of standard Maxwell’s BC. At the same time, the non-zero current normal component $j^{(0)}_g$ gives rise to discontinuity of the electric field component $E_\tau$, then the current tangential component $j^{(0)}_\tau$ leads to a jump in $B_\tau$.

The BC (9, 10) acquire practical significance if some constitutive equations are given, which indicate how the polarization current $j^{(0)}_\tau$ is generated under electric field of the electromagnetic wave at the point $z = \zeta$. In the case of normal incidence of light, $k_x = 0$, the field $E_\tau$ is continuous, i.e. $E_\tau (\zeta \pm 0) = E_\tau (\zeta)$, and discontinuity is experiencing (with $j^{(0)}_\tau \neq 0$) only the field $B_\tau$. Then $j^{(0)}_\tau \propto E_\tau (\zeta)$ and BC (10) takes the form

$$B_\tau (\zeta + 0) - B_\tau (\zeta - 0) = i\alpha (\omega) e_z \times E_\tau (\zeta)$$  \hspace{1cm} (11)

where $\alpha (\omega)$ is some proportionality coefficient, which is included in a constitutive equation for a singular current. The resonant nature of the singular current $j^{(0)}_\tau$ is governed by the corresponding frequency dependence of $\alpha (\omega)$ which can be set in a fairly general form, using the classical oscillator model as follows

$$\alpha (\omega) = \frac{2\omega \Gamma_{00} \sqrt{\varepsilon_b}}{\omega_b^2 - \omega^2 - i\omega \Gamma}$$  \hspace{1cm} (12)

where $\omega_0$ is the resonant frequency of the optical transition, $\Gamma_{00}$ is the radiative damping of the dipole oscillator layer, $\Gamma$ is the full non-radiative damping constant, $\varepsilon_b$ is the background (in the vicinity of $\omega_0$) dielectric constant. The representation of the coefficient $\alpha (\omega)$ in the form (12) corresponds, in
particular, to results of reflectance calculations performed for narrow quantum wells where 2D-excitons can be excited [1,3].

3. Numerical simulation of reflectance spectra
As an example of using the approach discussed in the present work, numerical calculations of reflectance spectra of light for a semiconductor structure consisting of CdTe monolayers embedded in a ZnTe semiconductor matrix are performed. Such a structure of a sufficiently high optical quality can actually be fabricated on GaAs substrates and used to study a wide class of phenomena associated with the optical excitation of planar monolayers. Our model calculations allow us to preliminarily estimate the geometrical and optical parameter values of a designed structure, which is necessary for purposeful controlling the growth processes of the objects under study.

Figure 1 schematically shows such a structure which includes \( N \) monolayers of CdTe located with respect to each other at a certain period \( d \). Monolayers (\( \delta \)-layers) are placed in a relatively thick bulk layer (of the thickness \( L > Nd \)) of ZnTe that covers a semi-infinite GaAs substrate. The nearest to the structure surface \( \delta \)-layer is moved at a distance \( d_{CAP} \) from it. Incident light comes from vacuum at an angle \( \theta \) in the \( xz \)-plane of incidence.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{structure.png}
\caption{The structure under study.}
\end{figure}

It should be noted that the spectral region of interest, where one expects to observe resonance effects due to \( \delta \)-layers, is located relatively close to the fundamental absorption edge of ZnTe. Therefore, the frequency dispersion of the matrix material permittivity may be significant. In addition, on the shortwave wing of the spectrum, spectral features due to exciton states should be expected.

To account for the spectral features associated with the ZnTe absorption edge, we used the approximating expression for the background permittivity \( \varepsilon_b(\omega) \) which was proposed in [9] to calculate the reflection spectra of multilayer systems based on ZnS,Se\(_{1-x}\) solid solutions:

\[ \varepsilon_b(\omega) = \varepsilon_{00} + \frac{\omega_p^2}{\omega_{exc}^2 - \omega^2 - i\omega \Gamma_{exc}} + \frac{\hbar \omega_p^2}{4 R_y \omega_g} \ln \left( \frac{\omega_{g+\omega+i\Gamma_g/2}}{\omega_{g-\omega+i\Gamma_g/2}} \right) \]  \hspace{1cm} (13)

In this expression (disregarding spatial dispersion) \( \varepsilon_{00} \) is the background dielectric constant, \( \omega_p \) is the “plasma” frequency of the lowest exciton resonance, \( R_y \) is the binding energy of this exciton, \( \omega_g \) is the frequency of the absorption edge, \( \Gamma_g \) is the damping constant (broadening) of the absorption edge, \( \omega_{exc} = \omega_g - R_y / \hbar \) is the resonance frequency of the exciton, \( \Gamma_{exc} \) is its decay constant. Choosing the values of these parameters, we were guided by reference data on ZnTe [10]:

\[ \varepsilon_{00} = 7.32 + 0.05i, \ \hbar \omega_p = 116 \text{ meV}, \ R_y = 12 \text{ meV}, \ \hbar \omega_g = 2390 \text{ meV}, \ \hbar \Gamma_g = 1.5 \text{ meV}, \ \hbar \Gamma_{exc} = 5.5 \text{ meV}. \]

For a GaAs substrate, its characteristic dielectric constant at the 535 nm wavelength was taken to be \( \varepsilon_s \approx 16.86 + 2.40i \) (see [11]). As for the parameters of the singular layer appearing in the formula (12), their values \( \omega_0 = 2281 \text{ meV}, \ \Gamma_{00} = 2.5 \text{ meV} \) and \( \Gamma = 10.5 \text{ meV} \) were selected from
considerations of the most descriptive representation of expected resonance effects. It should be noted that the choice of parameters for a singular layer is largely determined by the microscopic mechanism of the polarization excitation. In this paper, we do not specify such a model, while qualitatively focusing on the preliminary experimental data available at our disposal, relating to ZnTe samples with embedded ultrathin CdTe layers. The parameter values used in the calculations (especially $\Gamma_0$ and $\Gamma'$), in principle, may differ significantly from those typical of other known systems [1,6]. Apparently, in the context of our calculations, as the most adequate characteristic of singular optical resonance should be considered not directly the numerical values $\Gamma_0$ and $\Gamma'$, but their ratio $\Gamma_0/\Gamma'$. The main advantage of our approach is that it is fairly general for describing the resonant optical properties of sufficiently thin planar layers and is not limited to the specific microscopic model of the optical transition in such layers.

Figure 2(a) demonstrates the variation with the monolayer number $N$ ($N=1,3,5,10$) of the reflection spectrum for the resonant Bragg structure at normal ($\theta=0^\circ$) incidence of light. The resonant singular monolayers are placed with the spatial period of $d=98.2$ nm inside the thick matrix layer of the $L=2500$ nm thickness at the distance $d_{CAP}=198$ nm from the surface. It is clearly seen that at the selected $d$ value (corresponding to the condition $d \approx \lambda/2$ where $\lambda$ is the wavelength of light in the matrix material) there is a noticeable gain of reflectivity with increasing $N$ in the region of the resonant frequency $\omega_0 = 2281$ meV. The resonance peak of reflection is manifested against the background Fabry-Perot oscillations which converge and fade as they move to the long-wave tale of the fundamental absorption edge.

Figure 2(b) shows the spectra of the normal reflection of light for the Bragg structure with ten $\delta$-layers in relation to the thickness $d_{CAP}$ ($d_{CAP}=198, 162$ and $138$ nm) of the covering layer. As can be seen from the calculation, the spectrum can drastically change depending on the $d_{CAP}$ value.

Finally, figure 2(c) demonstrates the change in the reflection spectrum for $p$-polarization component as the angle of incidence increases ($\theta=0^\circ, 44^\circ, 54^\circ, 70^\circ$; $d_{CAP}=198$ nm). It is worth to note that at calculating the reflection coefficient for oblique incidence we supposed that $f_z^{(0)} = 0$ and $f_t^{(0)} \neq 0$, thereby assuming $E_t$-continuity at all points of the medium. It is important to emphasize here that at the angle of incidence $\theta=70^\circ$ close to the Brewster background angle ($\tan\theta \approx \sqrt{\varepsilon_b}$), the Fabry-Perot interference structure is substantially suppressed, which makes it possible to distinguish most clearly the contributions due to $\delta$-layers.
4. Conclusion
In the present work, a theoretical approach is developed to the analysis of the reflection spectra of light formed by semiconductor planar structures with embedded ultrathin layers that exhibit resonant optical properties. The approach is quite general and is based on a certain phenomenological parameterization of a layer microscopic structure using the formalism of singular generalized functions. The nature of the layer resonant properties can be quite different, including the specificity of narrow quantum wells for excitons, 2D excitons in monolayers, localized excitons in fairly densely located quantum dots forming a plane layer, planar defects bounding excitons, just molecular optical excitations, etc. The main limitation in using the approach is to fulfill the requirement that the thickness of the singular layer is small compared with the characteristic wavelength of light inside the structure. As a practically important example, calculations of the spectra formed by Bragg structures based on a ZnTe bulk crystal containing singular CdTe layers are performed in the work. Our model calculations make it possible to estimate the real geometric and optical parameters of the structures intended for the planned experimental studies.

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