

Surface Exciton Polariton Spectrum in Semiconductor Superlattices

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We report a surface exciton polariton spectrum in superlattices made up of two alternating media A and B, where one of the layers shows spatial dispersion in its frequency dependent dielectric function. We have used a theoretical approach based on the effective medium theory to determine the surface exciton polariton dispersion relation. The inclusion of an external magnetic field parallel to the interfaces of the superlattice is also considered, and it leads to the so-called magnetoplasmon-polariton excitations. It is shown that, in the absence of the external magnetic field, the surface modes are doubly degenerate, corresponding to propagation in opposite directions for a given magnitude of the wavenumber k_x . However, in the presence of the external magnetic field, this degeneracy is removed. We have also calculated the so-called Attenuated Total Reflectivity (ATR) to probe the surface exciton polariton modes. The effective-medium description has been widely used in ATR spectroscopy of semiconductor superlattices, and it generally gives a good account of the data together with a clear physical interpretation of the various spectral features.

I. Introduction

The study of the influence of exciton-polaritons (EP) as coupled-mode excitations made up from dipole-active excitons interacting with photons in the so-called excitonic region has been the subject of intensive study for many years. The main achievements of the EP description were postulated by Pekar^[1], Hopfield^[2], and Hopfield and Thomas^[3]. In their papers they proved that the finite wavevector dependence in the dispersion equation for the total energy of the exciton could play a decisive role in the optical properties of a crystal in the exciton energy region, leading to a significantly different behavior from the simpler case of phonon-polaritons. Indeed, the description of EP requires the inclusion of a spatial dispersion term βk^2 in the definition of the usual dielectric function, i.e.^[3]:

$$\epsilon(\omega, k) = \epsilon_\infty \left(\frac{\omega_L^2 - \omega^2 + \beta k^2}{\omega_T^2 - \omega^2 + \beta k^2 - i\omega\Gamma} - \frac{\omega_p^2}{\omega(\omega - i\Gamma)} \right) \quad (1)$$

where ϵ_∞ is the background dielectric constant, $\omega_L(\omega_T)$ and ω_p are the longitudinal LO (transverse TO) phonon

and plasma frequencies, respectively, and Γ is an empirical damping constant. Here β is the term responsible for the spatial dispersion, and is defined by $\beta = \hbar\omega_0/m^*$, with ω_0 being the frequency of the uncoupled exciton mode, which has an effective mass m^* .

The bulk EP dispersion relation is found by solving Maxwell's equations with the use of the dielectric function (1). This gives a longitudinal mode, defined by $\epsilon(\omega, k) = 0$, and a transverse mode, defined by $k^2 = \epsilon(\omega, k)\omega^2/c^2$. There are, therefore, two transverse modes which propagate in the same direction of the crystal and with the same polarization, at a given frequency ω . The most important qualitative change in the EP dispersion relation is that there is no stop band in the dispersion curve; at least one mode propagates for every frequency ω .

Suppose that an incident p -polarized light comes from vacuum to the surface excitonic medium which is parallel to the xy -plane. Two transverse modes, of wavevectors k_1 and k_2 , and a longitudinal mode of wavevector k_L are excited and can propagate in the medium. It is then necessary to evaluate the reflection

coefficient R_P , two “transverse” transmission coefficients T_1 , and T_2 , and a “longitudinal” transmission coefficient T_L . However, Maxwell’s equations provide only two boundary conditions. Therefore we should consider the so-called exciton’s additional boundary conditions (ABC), where most of them, proposed so far, can be cast in form (see [4] and the references therein):

$$\mathbf{P}(z) + \alpha \partial \mathbf{P}(z) / \partial z = 0 \quad \text{at the surface.} \quad (2)$$

Here $\mathbf{P}(z)$ is the excitonic polarization vector field in the spatially-dispersive crystal and α a phenomenological parameter. This model was successfully employed to calculate optical properties, like RBS (Resonant Brillouin Scattering), reflectivity and transmissivity of crystals in the excitonic region^[5]. Expressions for the reflection and transmission coefficients for any value of α can be found in [6].

It is the aim of this work to extend our theory presented in a previous paper^[7] to determine the EP dispersion relation that can propagate in superlattices, where one of the constituents media is excitonic. We consider also the presence of an external magnetic field \mathbf{B}_0 applied to the superlattice interfaces, along the y -direction. We use again the framework of the effective medium approach, which has been successfully employed in previous works^[7–11].

The plan of this paper is as follows: in section II we calculate the frequency-dependent effective dielectric tensor, taking into account the presence of the field as well as the fact that one constituent of the superlattice is excitonic. Then we determine the magnetoplasmon-polariton dispersion relation for the bulk and surface modes, for a particular choice of the ABC (3). As in the case of an ordinary magnetoplasmon-polariton^[12], the latter exhibits the property of reciprocity, that is, the mode frequency changes when the magnetic field is reversed. Section III is devoted to the discussion of attenuated total reflection (ATR), which is the major technique to study electromagnetically coupled surface excitations.

II. Dielectric tensor and dispersion relation

The semi-infinite superlattice structure, occupying the region $z > 0$, is composed alternating layers of materials A and B, one of which (medium A) is excitonic.

Layers A have thicknesses d_A and d_B , and dielectric functions $\epsilon_A(\omega, k)$ and $\epsilon_B(\omega)$ respectively, where $\epsilon_B(\omega)$ is given by (1) with $\beta = 0$. The cartesian axes are chosen in such a way that the z -axis is normal to the plane of the layers, and the structure is terminated at the plane $z = 0$, with the half-space $z < 0$ filled with a material that has a frequency-independent dielectric function ϵ_C .

Following the approach of [13,14], the effective dielectric tensor can be obtained from the relation of the averaged displacement vector $\langle \mathbf{D} \rangle$ and the averaged electric field $\langle \mathbf{E} \rangle$, where the average is over the unit cell of the superlattice. Considering that E_x and D_z are independent of z inside layer A, and making use of Maxwell’s boundary conditions together the ABC (eq. 3), the effective dielectric tensor can be given by:

$$\langle \ddot{\epsilon} \rangle = \begin{bmatrix} \epsilon_{xx} & 0 & -i\epsilon_{xz} \\ 0 & \epsilon_{yy} & 0 \\ i\epsilon_{xz} & 0 & \epsilon_{zz} \end{bmatrix} \quad (3)$$

where the elements of the dielectric tensor can be found elsewhere^[15].

Now assume that an electromagnetic wave, p -polarized (TM mode), is incident from outside medium onto the superlattice. Using equations of Huang type, we can set up the coupling of the excitonic polarization field $\mathbf{P}(z)$ to the macroscopic electromagnetic fields \mathbf{E} and \mathbf{H} (see [15] for details). Surface polaritons are free oscillations of the electromagnetic fields at an interface. Following [16, 17], they should not be forced by an incident wave, which means a reflectivity problem with zero incident field, i.e. a pole in the reflectivity. However, the reflectivity is given, in general by the well known formula^[18]:

$$R_p = \left| \frac{Z(\text{non-local}) - Z(\text{local})}{Z(\text{non-local}) + Z(\text{local})} \right|^2 \quad (4)$$

where Z , the surface impedance of the wave, is defined by:

$$Z(\text{local}) = E_x(0^-) / H_y(0^-) \quad \text{for } z < 0 \quad (5)$$

$$Z(\text{non-local}) = E_x(0^+) / H_y(0^+) \quad \text{for } z > 0 \quad (6)$$

Therefore, the surface magnetoplasmon-polariton dispersion relation is given by:

$$Z(\text{local}) + Z(\text{non-local}) = 0 \quad (7)$$

This dispersion relation is rather complicated, and it is difficult to draw general conclusions from it [15]. We believe that useful physical insight can be obtained if we take the limit of small non-local effects, mainly if one is more concerned with the importance of the roles played by different ABC's. Fortunately, for $\alpha = \infty$ it can be proved (see [19] for details) that the lowest-order non-local correction vanishes identically. It is then easy to show that in this approximation, the surface polaritons propagating perpendicular to the magnetic field \mathbf{B}_0 have their dispersion relation obtained for an effective medium described by a *k-independent gyrotropic dielectric tensor*, whose elements have been replaced by *the k-dependent one*, derived from (1). Its formal expression is [10]:

$$\left(\frac{\epsilon_{xx}}{\epsilon_{zz}} k_x^2 - \frac{\omega^2}{c^2} \epsilon_{\text{eff}} \right)^{1/2} + \epsilon_{\text{eff}} \left(k_x^2 - \frac{\omega^2}{c^2} \right)^{1/2} = k_x \frac{\epsilon_{xx}}{\epsilon_{zz}} \quad (8)$$

with

$$\epsilon_{\text{eff}} = \epsilon_{xx} - (\epsilon_{xz}^2 / \epsilon_{xx}) \quad (9)$$

Now, we present some numerical examples to illustrate the bulk and surface polariton spectra. In what follows, we assume physical parameters typical of electron concentration in GaAs-GaAlAs superlattices. The medium outside the superlattice is considered to be vacuum. We take thicknesses corresponding to $d_A = 40$ nm and $d_B/d_A = 2$. Also, we consider the following physical parameters in numerical calculations: $\epsilon_{\infty A} = 10.9$, $\epsilon_{\infty B} = 10.22$, $\omega_{LA} = 5.496$, $\omega_{TA} = 5.057$, $\omega_{LB} = 6.979$, $\omega_{TB} = 6.731$, and $\omega_{PA} = \omega_{PB} = 2.54$ (all the frequencies in units of 10^{13} Hz)

The applied magnetic field is measured through the cyclotron frequency ω_C , which is taken as the value of the plasma frequency. The parameter β , responsible for the spatial dispersion in medium A, is equal to 2.055×10^{13} Hz; the damping factor is taken to be zero in both media.

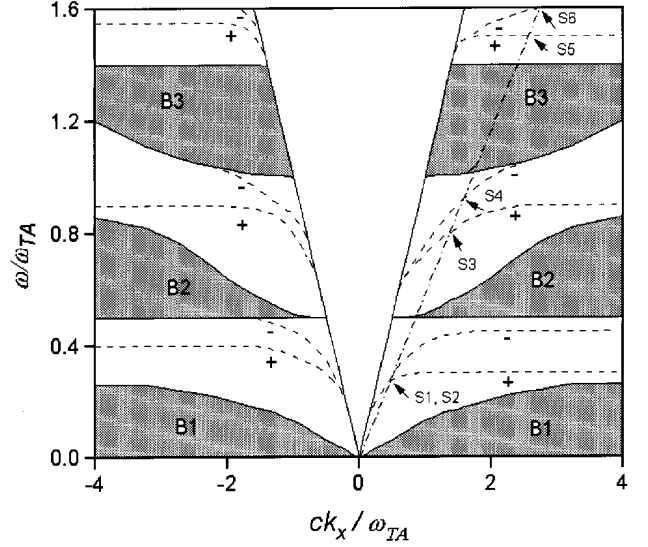


Figure 1. Surface-polariton spectra, *p*-polarization, within the effective-medium approach, for $\alpha = \infty$. The physical parameters used here are described in the text.

Fig. 1 shows the surface polariton dispersion curves given by (8), which is appropriate for the $\alpha = \infty$ case in the limit of small non-local effect. We have plotted a reduced frequency ω/ω_{TA} against a dimensionless wavevector ck_x/ω_{TA} . Here the dashed line means the ATR scan line; the vacuum line (full line in the figure) is defined by $k_x = \omega/c$. For completeness we have also presented the bulk bands, which are given by [10]:

$$\epsilon_{xx} k_x^2 + \epsilon_{zz} q_z^2 = (\epsilon_{xx} \epsilon_{zz} - \epsilon_{xz}^2) \omega^2 / c^2 \quad (10)$$

Here,

$$q_z^2 = (\omega/c)^2 \epsilon_{xx} - (\epsilon_{xx} / \epsilon_{zz}) k_x^2 \quad (11)$$

For a given ω the bulk modes may have any value of k_x , so they occupy the continuum region showed shaded in Fig. 1. Also these modes are reciprocal, that is, they are the same for $+k_x$ as for $-k_x$. On the other hand the surface polaritons, which are represented by dotted lines in Fig. 1, are, as expected, non-reciprocal. The points with the labels S1 to S6 are identified as being the crossing of the ATR scan line with these modes. Observe that the points S1, S2 are so close to each other that it is difficult to distinguish one from the other. All the modes start at their low frequency end on the vacuum light line.

It is possible to distinguish two different types of surface polaritons: those, which persist as $k_x \rightarrow \infty$ are the *real* surface modes, while the others, merging at the

high-frequency end into a bulk region at a finite value of k_x , are *virtual* surface modes. All the modes start at their frequency end on the vacuum light line. Observe that the degeneracy of the surface modes presented in [7] in Fig. 2 of is now removed by the applied magnetic field, leading to the modes labeled by the plus (+) and minus (-) signs in Fig. 1. They correspond to propagation in opposite directions for the wavevector \mathbf{k} .

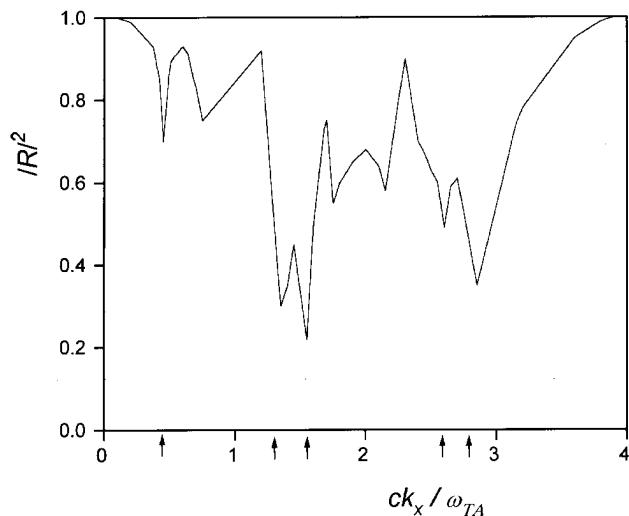


Figure 2. Calculated ATR spectrum for p -polarized incident light and the parameters given in the text.

III. ATR spectrum

The surface polariton modes can now be studied theoretically by means of ATR. The theoretical description of this method can be found in many papers on this subject (for a good account of the method see Ref. 20), and we simply quote the analytical result. For incidence plane xz and p -polarization, the ratio of the incident to the reflected energy within the framework of the effective medium approach is given by [21]:

$$|R|^2 = \left| \frac{Q \exp(-2iq_0 d) - (q_0 \epsilon_{xx} - q_z)(q_0 \epsilon_x + q_z)}{Q - (q_0 \epsilon_{xx} + q_z)(q_0 \epsilon_s + q_s) \exp(-2iq_0 d)} \right|^2 \quad (12)$$

where q_s , q_0 are the z -component of the wavevector in the prism (which is placed at a distance d above the superlattice in this technique) and the air, respectively, i.e. $q_j = [\epsilon_j(\omega/c)^2 - k_x^2]^{1/2}$ for $j = S$ or 0; also,

$$Q = ({}_0\epsilon_{xx} + q_z)(\epsilon_s q_0 - q_s) \quad (13)$$

The theoretical ATR spectrum is shown in Fig. 2 (for the particular value of $\alpha = \infty$). The dielectric constant of the prism and air is given by $\epsilon_s = 10$ (suitable for a Si prism). Also $\epsilon_0 = 1$ (vacuum), the thickness for the air-gap is equal to $5 \mu\text{m}$, and we use a damping factor corresponding to $4.0 \times 10^{-4} \omega_{pA}$. The angle of incidence θ_i , is chosen to be 20° , which is slight by greater than the critical value 17.1° for internal total reflection of a silicon-air interface.

The surface mode features in the ATR spectra, represented by the arrows, are here identified because their frequencies coincide with the points where the ATR angular scan line crosses the dispersion curve depicted in Fig. 1, as well as due to a convenient choice of the size the air gap. Of course, for smaller air gap, the surface polariton may be perturbed by the proximity of the prism, and one could not get all surface mode dips. Conversely, with large air gaps, the coupling of the evanescent wave to the superlattice is weak, and some dips may not brought out as clearly.

Besides the ATR method, two other important techniques may be mentioned^[22]. First the deposition of a grating on the specimen surface enables coupling of the incident radiation to the surface and guided modes; in general the increase of wavenumber k_x by $2n\pi/\lambda$ due to the grating period λ is relatively large, so that retardation effects are unimportant. Second, we can probe the surface polariton mode by oblique-incidence reflectivity. Although we have not presented any discussion regarding this technique, the algebraic expressions can be easily reached by taking the limit of the air-gap thickness d tending to zero in the expressions derived here. Furthermore, we can shown that the resulting spectra have no more information than those discussed in this paper.

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References

1. S.I. Pekar, Zh. Eksper. Teor. Fiz. **33**, 1022 (1957) (Sov. Phys. JETP **6**, 785 (1958)); *ibid.* Fiz. Tvers. Tela Leningrad **4**, 1301 (1962) (Sov. Phys. Solid State **4**, 953 (1962)).

2. J. J. Hopfield, Phys. Rev. **112**, 1555 (1958).
3. J. J. Hopfield and D.G. Thomas, Phys. Rev. **132**, 583 (1963).
4. J. L. Birman, in *Excitons*, eds. E. I. Rashba and M. D. Sturge (North Holland, Amsterdam, 1982).
5. C. Weisbuch and R. G. Ulbrich, in *Light Scattering in Solids III: Recent Results*, eds. M. Cardona and G. Güntherodt (Springer Verlag, Berlin, 1982).
6. E. L. Albuquerque and C.E.T. Gonçalves da Silva, J. Phys. **C 18**, 669 (1985); *ibid*, J. Physique **C5**, 61 (1984).
7. E.L. Albuquerque and P. Fulco, Phys. Stat. Sol. (b) **182**, 357 (1994).
8. N. S. Almeida and D. L. Mills, Phys. Rev. **B 38**, 6698 (1988).
9. N. S. Almeida and D. R. Tilley, Solid State Commun. **73**, 23 (1990).
10. M. C. Oliveros, N. S. Almeida and D. R. Tilley, Semicond. Sci. Technol. **8**, 441 (1993).
11. F. G. Elmznghi, N. C. Constantinou and D. R. Tilley, J. Phys.: Condens. Matter **7**, 315 (1995).
12. R. E. Carnley, Surf. Sci. Rep. **7**, 103 (1987).
13. V. M. Agranovitch and V. E. Kravstov, Solid State Commun. **55**, 85 (1985).
14. N. Raj and D. R. Tilley, Solid State Commun. **55**, 373 (1985).
15. E. L. Albuquerque and P. Fulco, Z. Phys. B (in press, 1995).
16. M. Cardona, Am. J. Phys. **39**, 1277 (1971).
17. B. B. Dasgupta and A. Bagchi, Phys Rev. B, **19**, 4935 (1979).
18. K. L. Kliewer and R. Fuchs, Phys. Rev. B, **172**, 607 (1968).
19. P. Halevi and R. Fuchs, J. Phys. **C17**, 3869 (1984); *ibid* 3889 (1984).
20. A. Otto, in: *Optical Properties of Solids, New Developments*, ed. B.O. Serafim (North Holland, Amsterdam, 1976).
21. E. L. Albuquerque and M. G. Cottam, Phys. Rep. **233**, 67 (1993).
22. T. Dumelow and D. R. Tilley, J. Opt. Soc. Am. **A 10**, 633 (1993).