Excitons in a spatiotemporal lattice

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A spatial superlattice becomes a spatiotemporal lattice and the minibands (MB's) change to quasienergy MB's under intense laser pumping. The width of the quasienergy MB collapses at a particular ratio of the field to frequency of the driving laser. It is shown that both the excitonic binding energy and the absorption oscillator strength reach maxima at this particular critical ratio. Optical studies may, therefore, lead to a verification of the laser-induced miniband collapse.

The advent of sophisticated techniques for the growth and characterization of ultrathin layers of semiconducting materials has made it possible to fabricate superlattices (SL's) with accurately controlled compositions and widths of the individual layers.[1,2] Such a structure produces an artificially created periodic lattice superposed on the host lattice. This necessarily leads to the formation of bands of allowed and forbidden energies, called the minibands (MB's), in a similar fashion to what happens in crystals. The widths of the MB's may, at the most, be a few meV only. The narrowness of these MB's gives rise to several nonlinear electrical and optical properties which make these structures useful in device applications.[1-5]

The response of a periodic superlattice to coherent radiation has been the subject of investigation since 1976.[6-10] While Ignatov and Romanov[6] calculated the current in a SL driven by a monochromatic laser from classical kinetic equations and showed that the current may reach zero for certain laser fields, a truly quantum-mechanical model valid in the nonperturbative regime is due to Holthaus and co-workers.[8-10] The authors predicted that at certain values of the field-to-frequency ratio of the driving laser there will be a complete collapse of the MB's. Indications for such an MB collapse, according to these authors, may be found by measuring electronic transport properties in the SL and also by observing intraminiband absorption. As far as the authors are aware no such verification has been reported in the literature. In this paper we propose an alternate scheme for verifying this concept by using interband optical absorption. Since it is established that the excitonic processes dominate the band-edge optical response of quantum confined structures, we find it important to explore the effect of electron-hole Coulomb interaction in the present system. This paper reports a theoretical treatment of the excitonic resonance in a periodic SL in the presence of an intense laser pump having much smaller energy than the inter-MB gap. We have explored the response of such a system to a probe beam which is tuned to the excitonic band gap. The main conclusion that emerges is that the excitonic MB collapses in a laser-driven SL and that there is a maxima in the absorption oscillator strength to register the collapse associated with a blueshift of the absorption threshold. The scheme is thus an optical analog of that used to observe the Wannier-Stark localization (WSL) in a SL.[11-13]

The Hamiltonian for an electron in a SL exposed to a laser polarized along the growth direction (z) is

$$H_0(z,t)=-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2}+V_{\mathrm{SL}}(z)-eF(t)z \ , \tag{1}$$

where $V_{\mathrm{SL}}(z)$ is the SL model potential, e is the electronic charge, and $F(t)$ is the electric field associated with the laser:

$$F(t)=F_0\sin(\omega_{\mathrm{IR}}t) \ , \tag{2}$$

ω_{IR} being the angular frequency of the infrared (IR) pump laser.

The Hamiltonian is periodic in both space and time due to the periodicity in $V_{\mathrm{SL}}(z)$ and $F(t)$:

$$H_0(z,t)=H_0(z+L,t)=H_0(z,t+T) \ , \tag{3}$$

where $T=2\pi/\omega_{\mathrm{IR}}$ and L is the period of the SL. We also note that both the spatial and temporal translation operators commute with the Hamiltonian $H_0(z,t)$. It has been pointed out by Holthaus and co-workers[8-10] that such a system is described by spatiotemporal Bloch functions instead of the usual spatial Bloch functions. They have also pointed out that this system should behave as a new object with its own quasienergy dispersion relation.

The electronic states without the Coulomb interaction may be expressed as

$$\psi(z,t)=\exp\{i(kz-\xi t)\}\phi(z,t) \ , \tag{4}$$

where $\phi(z,t)$ is periodic in both space and time and k and ξ are the quasimomentum and quasienergy, respectively. These states play the role of the stationary states in a quantum system having both spatial and temporal periodicities.

The fact that the vector potential $A(t)$ is independent of z leaves k a good quantum number. An additional condition that $\hbar\omega_{\mathrm{IR}} \ll$ inter-mini-band-gap is set so that the adiabatic approximation remains applicable. The spatiotemporal Bloch waves may, then, be written as[9,10]

$$\psi_{nk}(z,t)=\exp\left\{ikz-i\hbar^{-1}\int d\tau\, E_n[k-eA(\tau)/\hbar]\right\}\times\phi_{nk}(z,t) \ , \tag{5}$$

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where $E_n(q)$ is the functional form of SL dispersion relation.

For a periodic field as expressed in Eq. (2) spatiotemporal Bloch waves assume the form as in Eq. (4) where the quasienergies are given by

$$\xi_{nk} = \frac{1}{\hbar T}\int_0^T d\tau\, E_n[k - eA(\tau)/\hbar] \; . \tag{6}$$

Under the nearest-neighbor tight-binding approximation, which is a very good approximation for a narrow MB SL, the quasienergy dispersion relation is obtained as

$$\xi_{nk} = \xi_{0n} - (\Delta_n/2\hbar)\cos(kL) J_0(eF_0L/\hbar\omega_{\mathrm{IR}}) \; . \tag{7}$$

The appearance of the zeroth Bessel function in Eq. (7) is an interplay between the spatial and temporal periodicities in the system. It can be easily noted that the effective widths of the quasienergy minibands are proportional to $J_0(eF_0L/\hbar\omega_{\mathrm{IR}})$ and hence they go to zero at the zeros of the Bessel function. This implies zero-group velocity of electrons which in turn signifies localization.

We now turn our attention to the excitons in a periodically driven SL. The Hamiltonian may be expressed as

$$H(z,t) = -\frac{\hbar^2}{2m_e}\frac{\partial^2}{\partial z_e^2} - \frac{\hbar^2}{2m_h}\frac{\partial^2}{\partial z_h^2} + V_{\mathrm{SL}}(z_e) + V_{\mathrm{SL}}(z_h) - eF(t)(z_e - z_h) - e^2/4\pi\varepsilon|r_e - r_h| \; . \tag{8}$$

In writing the Hamiltonian the in-plane (along the layer plane) center-of-mass motion is separated as usual. The Hamiltonian without the electric field has been solved by a few workers for SL's.[11,14,15] In essence, the excitonic MB is formed due to the presence of the periodic potential along the z direction. An excitonic reduced mass $\mu_z(0)$, totally different from the in-plane reduced mass, may be defined using the effective masses of electrons and holes along the z direction derived from the MB dispersion relations. The SL, therefore, behaves as an effective anisotropic medium,[14,15] the anisotropic factor tending to zero for the purely 2D system. An approximate scheme has recently been developed[16–20] to describe a switch over from a SL to a multiple quantum-well (QW) structure by introducing a fitting parameter η. This empirically deduced dimensionality parameter η is 3 for isotropic bulk medium and is 2 for a purely two-dimensional system, while the intermediate cases are described by a fractional dimensionality.

In considering the effect of the dynamic laser field on the excitonic behavior, we assume that the lifetime τ of the excitons are much larger than the time period T of the driving laser field. The available data for quantum confined structures[11] indicate that the excitonic lifetime is about a few ps. This sets a lower limit for the frequency of the driving laser. When $\tau \gg T$ ($\equiv 2\pi/\omega_{\mathrm{IR}}$), time can be taken as a continuum even if it is measured in the units of a laser time period. This condition then allows us to define a new effective mass $\mu_z(F)$ which takes care of both the spatial as well as the temporal periodicities. The excitonic reduced mass can, therefore, be derived from the quasienergy MB dispersion relations for electrons and holes which are given by Eq. (7). The medium, in this case, is strongly anisotropic and the extent of anisotropy can be altered by changing the laser field. The anisotropic effective-medium model used in Refs. 14 and 15 may be used over a large range with the exception that this model breaks down at the extreme anisotropy. We note that the periodically time-dependent Hamiltonian as in Eq. (8) may be reduced to a time-independent one by absorbing the periodic temporal variation into properly defined effective masses for both electrons and holes. We first calculate the binding energy of excitons in a spatiotemporal lattice by using the anisotropic effective-medium model. A conclusion derived from this model is that the binding energy is determined by the anisotropy parameter $\gamma = \mu_\parallel/\mu_z$. The reduced masses μ_z are determined from the MB dispersion relations for electrons and holes. Since the values of $\mu_\parallel$ remain unaffected by the laser field, it is only the value of μ_z that is to be evaluated. A plot of binding energy as a function of y ($=eF_0L/\hbar\omega_{\mathrm{IR}}$) given in Fig. 1 shows an increase with laser field. An increase in binding energy is associated with an increase in absorption. However, a calculation of absorption needs the knowledge of an exciton envelope function for different laser fields, that may be obtained through lengthy numerical work. Since our interest is only in illustrating the idea, we make use of the already established expression for opacity in terms of the abovementioned empirical dimensionality parameter η.

In a medium of dimension η the Wannier equation for excitons may be written as[17–20]

$$\left[-\frac{\hbar^2}{2\mu}\nabla_\eta^2 - \frac{e^2}{4\pi\varepsilon r}\right]\psi(r,\theta) = E\psi(r,\theta) \; , \tag{9}$$

where ∇_η^2 is defined as[16]

$$\nabla_\eta^2 = \frac{1}{r^{\eta-1}}\frac{\partial}{\partial r}\left[r^{\eta-1}\frac{\partial}{\partial r}\right] - \frac{L^2}{\hbar^2 r^2} \; , \tag{10}$$

with angular momentum operator L^2 defined as

$$L^2 = -\frac{\hbar^2}{\sin^{\eta-1}(\theta)}\frac{\partial}{\partial\theta}\left[\sin^{\eta-2}(\theta)\frac{\partial}{\partial\theta}\right] \; . \tag{11}$$

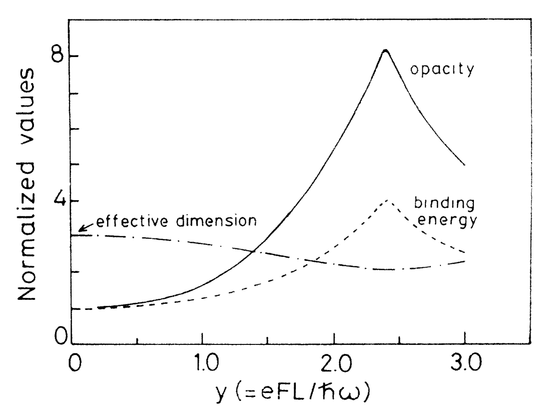

FIG. 1. Normalized values of opacity (——), excitonic binding energy (— — —), and effective dimension (—·—) versus y ($\equiv eFL/\hbar\omega$); L is the superlattice period and F and ω are the field and frequency of the driving infrared laser, respectively.

In Eq. (9), μ is the exciton-reduced effective mass derived from the quasienergy MB dispersion relations, ε is the effective dielectric constant, and r $(0<r<\infty)$ and θ $(0<\theta<\pi)$ are the pseudodistance and pseudoangle, respectively.[16]

Equation (9) can be solved by separating the variables to obtain the θ solutions in terms of the Gegenbauer polynomials while the r solutions (for the bound states) are given by the Laguerre polynomials. Since in this paper we are interested in the extent of variation in the excitonic absorption, we would like to focus on the corresponding spectral opacity of the medium which in a fractional dimensional space may be expressed as[20]

$$O_{\eta,n}(\hbar\omega)=A(\eta)\frac{2^{2\eta-4}}{\pi^{(\eta+1)/2}a_B^{\eta}}\frac{\Gamma^2(\eta/2)\Gamma((\eta-1)/2)}{\Gamma^9(\eta-1)} \times\frac{\Gamma(n+\eta-2)}{(n-1)!\left[n+\frac{\eta-3}{2}\right]^{\eta+1}}\delta(\hbar\omega-E_n)\ , \quad (12)$$

where η is a function of the laser field in the present case, a_B is the undriven value of the exciton Bohr radius in the SL, ω is the angular frequency of the probe laser which is being absorbed, Γ is the Euler's gamma function, and $A(\eta)$ is as defined in Ref. 20.

A simple method for the evaluation of the dimensionality factor η is discussed in Ref. 19. The effect of the laser field on η is introduced through the parameter γ which is defined in that paper as $\mu_z(0)/\mu_z(F_0)$. In the present context γ may be expressed as

$$\gamma=J_0^{-1}(eF_0L/\hbar\omega_{\mathrm{IR}})\ . \quad (13)$$

γ, therefore, goes to zero at the zeros of the Bessel function thereby making the system effectively a two-dimensional one $(\eta=2)$ while without the laser it remains a strongly anisotropic three-dimensional (3D) medium.

In Fig. 1 we have plotted the ratio of the opacity values in the presence of the infrared laser to that in its absence against y $(=eF_0L/\hbar\omega_{\mathrm{IR}})$. It shows a monotonic increase in the excitonic absorption with y, i.e., with increasing laser field for a constant frequency until y reaches the first zero of the Bessel function when the absorption reaches a peak. It is found that there is an increase of absorption by a factor of about 8.5 in the event of MB collapse. In our calculation the effect of broadening of states due to various homogeneous and inhomogeneous processes has not been included. The effects remain present both with and without the laser. There are indications[21,22] that excitonic peaks are more sharp in two dimensions than in a SL. The peak in Fig. 1 will, therefore, be enhanced further if broadening is included. Quenching of the first MB leads to a blueshift in the absorption threshold. The effective dimension η of the system as calculated in the present work is also plotted in Fig. 1. It appears as expected that the dimension becomes 2 when the MB collapses showing a peak in the opacity and the binding energy.

It may be mentioned at this point that a similar dimensional crossover with a SL at one end and a quantum well at the other may be accomplished by applying a dc electric field.[11–13] The effect named as Wannier-Stark localization is associated with a rise in the absorption oscillator strength and a blueshift in the absorption threshold. Some optoelectronic device applications[22,23] of WSL have already appeared in the literature. The present concept of MB collapse is thus an optical analog of WSL. Apart from testing the concept, device application may be envisioned if experimental work is pursued in this area.

It may be of interest to compute the values of laser frequency ω_{IR} and the power needed to observe the effect. We take any common system like GaAs/$\mathrm{Al_1Ga_{1-x}As}$ and take $\lambda_{\mathrm{IR}}=100$ μm to give $\hbar\omega_{\mathrm{IR}}=12.4$ meV. The photon energy must be less than the inter-MB separation. With $L=14$ nm, the needed value of the electric field is 2.2×10^6 V/m which requires that the intensity should be 63×10^8 W/m^2. Assuming minimum spot diameter ≈100 μm $(=\lambda_{\mathrm{IR}})$ this requires a laser with about 63 W of output.

In conclusion, a theoretical model is presented for the exciton problem in a periodic semiconductor superlattice in the presence of a strong infrared laser whose energy is much less than the inter-mini-band-gap. It is noticed that the absorption increases as the field to frequency ratio of the laser is increased and attains a maximum as a particular value is reached, which is the signature of the laser-induced collapse of minibands. There is a similar behavior of the excitonic binding energy. Some device applications of this phenomenon are alluring, but the experimental verification of the phenomenon itself might stand in its own merit.

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[1]L. Esaki, J. Phys. (Paris) Colloq. **48**, C5-1 (1987) and references therein.
[2]C. Weisbuch and B. Vinter, *Quantum Semiconductor Structures* (Academic, New York, 1991).
[3]L. Esaki and R. Tsu, IBM J. Res. Dev. **14**, 61 (1970).
[4]R. Tsu and L. Esaki, Appl. Phys. Lett. **19**, 246 (1971).
[5]F. Capasso, S. Sen, F. Beltram, and A. Y. Cho, in *Physics of Quantum Electron Devices*, edited by F. Capasso (Springer-Verlag, Berlin, 1990), p. 181.
[6]A. A. Ignatov and Yu. A. Romanov, Phys. Status Solidi B **73**, 327 (1976).
[7]J. F. Lam, B. D. Guenther, and D. D. Skatrud, Appl. Phys. Lett. **56**, 773 (1990).
[8]M. Holthaus, Phys. Rev. Lett. **69**, 351 (1992).
[9]M. Holthaus and D. Hone, Phys. Rev. B **47**, 6499 (1993).
[10]M. Holthaus, Z. Phys. B **89**, 251 (1992).

[11]S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Adv. Phys. **38**, 89 (1989).
[12]J. Bleuse, G. Bastard, and P. Voisin, Phys. Rev. Lett. **60**, 220 (1988).
[13]E. E. Mendez, F. Agullo-Rueda, and J. M. Hong, Phys. Rev. Lett. **60**, 2426 (1988).
[14]M. F. Pereira, Jr., I. Galbraith, S. W. Koch, and G. Duggan, Phys. Rev. B **42**, 7084 (1990).
[15]Partha Ray and P. K. Basu, Phys. Rev. B **47**, 15 958 (1993).
[16]F. H. Stillinger, J. Math. Phys. **18**, 1224 (1977).
[17]X. F. He, Phys. Rev. B **43**, 2063 (1991).
[18]P. Lefebvre, P. Christol, and H. Mathieu, Phys. Rev. B **46**, 13 603 (1992).
[19]H. Mathieu, P. Lefebvre, and P. Christol, J. Appl. Phys. **72**, 300 (1992).
[20]P. Lefebvre, P. Christol, and H. Mathieu, Phys. Rev. B **48**, 17 308 (1993).
[21]Partha Ray and P. K. Basu, Phys. Rev. B **46**, 13 268 (1992).
[22]G. R. Olbright, T. E. Zipperian, J. Klem, and G. R. Hadley, J. Opt. Soc. Am. B **8**, 346 (1991).
[23]F. Devaux, E. Bigan, M. Allovon, J. C. Harmand, P. Voisin, M. Carre, F. Huet, and A. Carenco, Electron Lett. **28**, 48 (1992).