Dynamics of directly created excitons in asymmetric double quantum wells

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In this work, we have studied the dynamics of an electrically pumped exciton that is directly created by hole-assisted electron resonant tunneling in an asymmetric coupled quantum well system. We have numerically integrated in space and time the effective-mass Schrödinger equation for the electron-hole pair taking into account the two-particle nature of the tunneling process. We have also included the time evolution of the transition from spatially indirect to direct excitons. Considering a different exciton binding energy for each quantum well, we have obtained a different charge oscillation semiperiod for each binding energy case. In this way, the possibility is shown of having a new kind of terahertz electromagnetic radiation emerging from such a semiconductor quantum well system after electric pumping. [S0163-1829(96)06748-3]

I. INTRODUCTION

In recent years, semiconductor quantum wells have shown many new optical and electronic properties,[1] playing a new role in many electronic devices such as field-effect transistors, photodetectors, and quantum well lasers.[1,2] Recently, $GaAs/Ga_xAl_{1-x}As$ semiconductor coupled quantum wells have been used to observe tunneling charge oscillation in solids.[3] In such an experiment, the superposition of both symmetric and antisymmetric quantum well eigenstates in the conduction band led to coherent tunneling between both quantum wells, and thus to an electron-hole pair with a time-dependent distance. In this way, a time-varying excitonic dipole moment in the wells was obtained, so that it allowed the emission of electromagnetic radiation at the oscillation frequency.

There is also a great interest in other related phenomena, such as spatially coherent quantum well (QW) excitons, with an in-plane momentum $\mathbf{k}_{ex\|}\sim 0$, due to their capability of radiating in the perpendicular direction to the QW. Up to the present, resonant optical pumping was needed to create efficiently such excitons. In a recent experiment and theory, Cao *et al.*[4] have shown the possibility of the direct creation of $\mathbf{k}_{ex\|}\sim 0$ electrically pumped QW excitons. It seems interesting to extend the analysis to asymmetric coupled quantum wells (ACQW's). In the direct generation of excitons electrons tunnel from a *n*-type material through a barrier to the left quantum well (LQW). Because of the structure is closed by a *p*-type material, the process is assisted by the electron-hole Coulomb interaction, obtaining a two-particle process. We assume that holes thermally diffuse into the right quantum well (RQW). Under resonant conditions between two adjacent wells with different widths, electron-in-exciton's (the exciton electrons) can tunnel back and forth from one well to the other. Thus, we can expect that the generated time-dependent dipole can radiate THz electromagnetic waves before radiative recombination.

The process is as follows: First, the electron tunnels to the wide LQW forming a cross or spatially indirect (interwell) exciton with the hole in the narrow RQW. Second, if the resonant conditions for the exciton tunneling between both wells coincides with the electron resonant conditions between the free electron state in the *n*-type material and the electron-in-exciton state in the left well, (i.e., both resonances occur at the same applied field) the electron-in-exciton tunnels to the RQW, forming a spatially direct (intrawell) exciton. The difference with respect to Ref. 4 is that, in our work, the initial state is the free electron in the *n*-type GaAs layer and the hole in the RQW. The intermediate state is the cross or indirect exciton. If this exciton and the direct exciton are aligned, the final state will be the direct exciton with the electron and hole located in the RQW. Then, the electron-in-exciton oscillates between the wells before recombining. Finally, it will be shown that the dynamics of a directly created electron-hole pair will lead to a new type of time-dependent dipole moment in the system, and thus, a new kind of terahertz emission will emerge from the device.

II. METHOD OF CALCULATION

Our task is the analysis of these time-dependent excitonic processes in asymmetric double quantum wells, examining the dynamical evolution of excitonic electron-hole pairs.[5] The method of calculation will be based on the discretization of space and time for the carrier wave functions.

We simulated the injected-free electron, localized initially in the *n*-doped material, by a Gaussian wave packet. The hole wave packet stands initially in the RQW. After crossing the first barrier, the electron is trapped by the Coulomb interaction and forms the cross exciton with the hole. Under resonant conditions the electron-in-exciton packet is a linear superposition of the two eigenstates in the conduction band, and hence, nonstationary. Subsequently the electron-in-exciton packet will oscillate between the left and right wells. Meanwhile, the hole packet will remain localized in the right well. Besides this, we have two different exciton binding energies[6,7] that affect the wave-packet dynamics. As will be seen the cross-exciton binding energy is lower than the direct exciton binding energy.

We will consider in this work the ACQW potential represented in Fig. 1. The left electrode is a *n*-doped GaAs layer. A 100-Å $Ga_{0.65}Al_{0.35}As$ layer separates the electrode and the left QW. The left (80-Å) and right (58-Å) wells consist of GaAs. The barrier between the wells consists of a 25-Å-wide

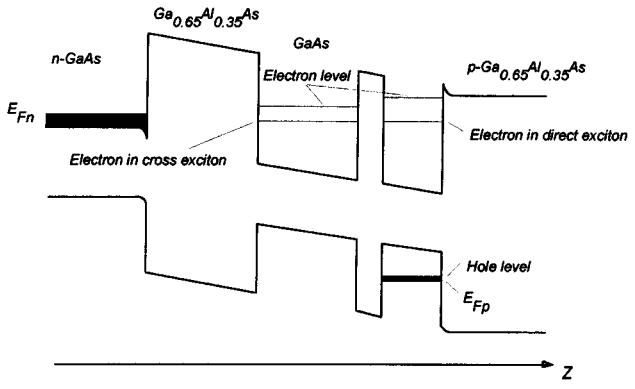

FIG. 1. Asymmetric double quantum well under electron-in-exciton resonant condition.

$Ga_{0.65}Al_{0.35}As$ layer. The right electrode is a p-doped $Ga_{0.65}Al_{0.35}As$ layer. In principle, in our proposed ACQW system, we have chosen a right quantum well width that allows electron-in-exciton coherent tunneling between the wells in the conduction band at the same applied field that is required for the free electron resonance from the n-doped electrode to the electron-in-exciton state in the left well. In order to simulate a realistic experimental structure, the n layer has a donor concentration of 4×10^{16} cm^{-3} and the p layer has an acceptor concentration of 1×10^{18} cm^{-3}. As has been stated before, holes diffuse thermally into the RQW. We have assumed a $m_e^*=0.067m_0$ effective mass in the conduction band and a $m_h^*=0.7m_0$ heavy-hole mass.

In order to study the dynamics of tunneling, we need to solve the time-dependent Schrödinger equation associated with the Hamiltonian for a spinless exciton in the heterostructure region. The excitonic wave function Φ is given by

$$\Lambda\Phi(\mathbf{r}_e,\mathbf{r}_h)=i\hbar\frac{\partial}{\partial t}\Phi(\mathbf{r}_e,\mathbf{r}_h), \tag{1}$$

where we have

$$\Lambda(\mathbf{r}_e,\mathbf{r}_h)=\Delta(\mathbf{r}_e,\mathbf{r}_h)+\sum_{i=e,h}\left[-\frac{\hbar^2}{2m_i^*}\frac{\partial^2}{\partial z_i^2}+V_i(z_i)\right] \tag{2}$$

and the operator Δ,

$$\Delta(\mathbf{r}_e,\mathbf{r}_h)=-\frac{\hbar^2}{2\mu_{xy}}\nabla_{xy}^2+V_C(\rho,z_e-z_h), \tag{3}$$

where the subscripts e,h refer to electrons or holes, respectively, and $V_e(z_e),V_h(z_h)$ are the quantum well potentials. The m_e^* and m_h^* values are the effective masses, μ_{xy} is the reduced $x-y$ plane electron-hole mass and $\rho=\rho_e-\rho_h$ is the relative motion within the quantum well plane. Dismissing the difference in the dielectric constants for well and barrier, the Coulomb potential is

$$V_C(\rho,z_e-z_h)=\frac{e^2}{\epsilon\sqrt{\rho^2+(z_e-z_h)^2}}. \tag{4}$$

We separate the total wave function Φ into the motion along z and the in-plane motion of the exciton $\phi(\rho)$,

$$\Phi=\Psi(z_e,z_h)\phi(\rho), \tag{5}$$

while retaining Coulomb effects in the growth direction:

$$\left\{\sum_{i=e,h}\left[-\frac{\hbar^2}{2m_i^*}\frac{\partial^2}{\partial z_i^2}+V_i(z_i)\right]+W(z_e-z_h)\right\}\Psi(z_e,z_h)$$
$$=i\hbar\frac{\partial}{\partial t}\Psi(z_e,z_h). \tag{6}$$

The potential

$$W(z)=\int_0^\infty d\rho\rho\left[\frac{\hbar}{2\mu_{xy}}[\partial_\rho\phi(\rho)]^2-\frac{e^2}{\epsilon\sqrt{\rho^2+z^2}}\phi^2(\rho)\right] \tag{7}$$

will be added to the heterostructure potentials.[8] This two-variable Schrödinger equation can be simplified introducing the factorization $\Psi(z_e,z_h)=\psi(z_e)\psi(z_h)$, thus obtaining the equations

$$\left[-\frac{\hbar^2}{2m_e^*}\frac{\partial^2}{\partial z_e^2}+V_e(z_e)+W_h(z_e)-eFz_e\right]\psi_e(z_e)$$
$$=i\hbar\frac{\partial}{\partial t}\psi_e(z_e), \tag{8}$$

$$\left[-\frac{\hbar^2}{2m_h^*}\frac{\partial^2}{\partial z_h^2}+V_h(z_h)+W_e(z_h)+eFz_h\right]\psi_h(z_h)$$
$$=i\hbar\frac{\partial}{\partial t}\psi_h(z_h), \tag{9}$$

that have to be solved together since the term

$$W_i(z)=\int dz'\,\psi_i^2(z')W(z-z') \tag{10}$$

couples ψ_e and ψ_h where $i=e,h$. To simplify this calculation we have used the ansatz[9] $\phi(\rho)=(2/a)e^{-\rho/a}$ for the in-plane motion of the exciton, where a corresponds to the Bohr radius in the two-dimensional case.

Now we discretize time by a superscript n and spatial position by a subscript j and k for the conduction and valence band, respectively. Thus, $\psi_e\rightarrow\kappa_j^n$ and $\psi_h\rightarrow\varphi_k^n$. The various $z_{e,h}$ values become $j\delta z$ and $k\delta z$ in the conduction and valence bands, where δz is the mesh width. Similarly, the time variable takes the values $n\delta t$, where δt is the time step. In this way, to treat the time development we have used an unitary propagation scheme for the evolution operator in both conduction and valence bands[10,11] obtaining a tridiagonal linear system that is solved by standard numerical methods. We have assumed that a mixed electron-hole pair is initially created at $t=0$. Then, Eqs. (6) and (7) are numerically solved using a spatial mesh size of 0.5 Å and a time mesh size of 1 fs and a finite box (2000 Å) large enough to neglect border effects.

At certain electric field, electron-in-exciton energy levels of the right and left wells are aligned in the conduction band while, in the valence band, the hole level is mainly localized in the right well. Then, the electron charge density will oscillate in a band with a certain tunneling period, and the hole

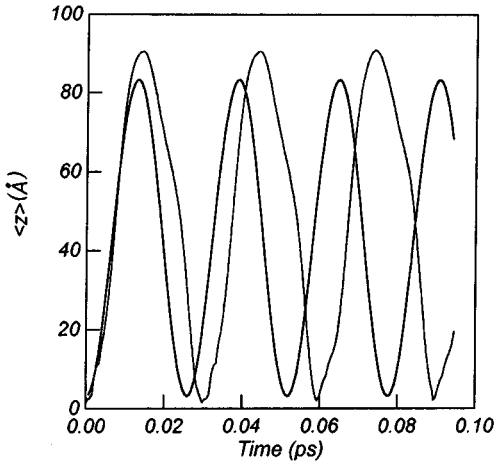

FIG. 2. The averaged z value of the electron position. Black line: free electron; gray line: electron-in-exciton.

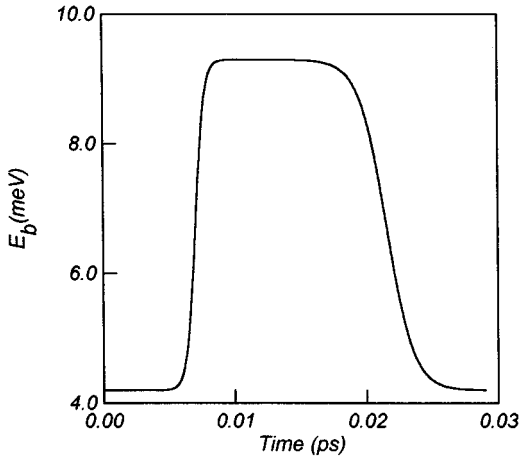

FIG. 3. Exciton binding energy versus time during a period. The initial state corresponds to the spatially indirect exciton in the left well.

will remain localized. We have neglected the mixing effects in the valence band.[12] Such an approximation is well justified for our structure potentials.[5]

The numerical integration in time allows us to obtain the average electron and hole positions $\langle z\rangle_{ab}^{e,h}$ and the carrier charge density $Q_{ab}^{e,h}$ in a defined semiconductor region $[a,b]$ at any time t,

$$\langle z\rangle_{ab}^{e,h}(t)=\int_a^b dz_{e,h}|\psi^{e,h}(z_{e,h},t)|^2 z_{e,h}, \tag{11}$$

$$Q_{ab}^{e,h}=\int_a^b dz_{e,h}|\psi^{e,h}(z_{e,h},t)|^2. \tag{12}$$

Since $z^{e,h}$ depend on t, the Coulomb interaction, which mostly determines the exciton binding energy together with the in-plane kinetic term, will depend on t too. To calculate the binding energy $E_b(t)$ we proceed as follows: at each t and $\langle z\rangle_{ab}^{e,h}(t)$ values $V_C(\rho,z_e-z_h,t)$ and $W_h(z_e,t)$ are assessed. In this way, the exciton binding energy is given by[13]

$$\begin{aligned}E_b(t)=&\frac{\hbar^2}{2\mu_{xy}a^2}-\frac{4e}{a^2}\int_{z_e}dz_e\psi_e^2(z_e,t)\int_{z_h}dz_h\psi_h^2(z_h,t)\\&\times\int_0^\infty\rho d\rho V_C(\rho,z_e,z_h,t)e^{-2\rho/a}\\=&\int_{z_h}dz_h\psi_h^2W_e(z_h,t).\end{aligned} \tag{13}$$

Other contributions to the binding energy are very small. In addition, E_b can also be calculated by minimizing the expectation value E of the total Hamiltonian Λ with respect to the parameter a at each step. Also, such a method allows us to obtain the time evolution of $\langle\rho\rangle=\langle\Phi|\rho^2|\Phi\rangle^{1/2}$.

III. RESULTS AND DISCUSSION

We first show the evolution of the electron-in-exciton averaged position, as well as the evolution of its charge density. In Fig. 2, we have plotted $\langle z\rangle_{ab}^{e}$ versus time, a and b being the left quantum well limits. One clearly sees the carrier oscillations for the electron-in-exciton packet when the free electron is placed initially in the left well (i.e., $z=0$, the center of the left well, at $t=0$). The oscillation period can be easily determined by analyzing the position of the oscillation peaks in the curves of Fig. 2. This figure also represents superimposed the free electron oscillations. Two perceptible differences exist between the motion of the two kinds of electrons because of the Coulomb interaction. First, the electron-in-exciton oscillations have a more remarkable asymmetry than the free electron oscillations. Second, the period of the former electron is about 10% greater than that of the latter (29 fs and 26 fs, respectively) in agreement with the calculations of Mohaidat *et al.*[14] The electron is accelerated when moving toward the hole and attracted when moving away from the hole. Both effects on the exciton wave packet almost compensate for each other and the net effect is a little increase of the oscillation period. On the other hand, the transition from spatially indirect to spatially direct exciton varies the Coulomb interaction and the in-plane kinetic energy. The exciton binding energy grows abruptly, from 4.2 meV to 9.1 meV, when the spatially indirect exciton changes to the direct one as shown in Fig. 3.[13] It should be noted that another effect on the exciton binding energy is attributable to the external electric field. When the electron and hole are spatially separated in the presence of an electric field, the Coulomb potential energy between them is increased while the in-plane kinetic energy is reduced, leading to the reduction of the binding energy and the increase of the spatial extension. Because of the above effects, there is a slight asymmetry in the oscillation shape. These features are also perceptible in the electron charge density oscillations $Q_{ab}^{e}(t)$ represented in Fig. 4. Thus, we have an oscillating electron-hole distance, whose amplitude is also oscillating in time in a different way from that of the resonant free electron or the electron trapped only by the Coulomb interaction.

It is worth mentioning other effects that contribute to the detected charge oscillations in experiments. First, the electron-in-exciton interaction with the existing phonons at finite temperature. The electron-optical phonon scattering reduces the mean life of the direct created exciton[15] and the

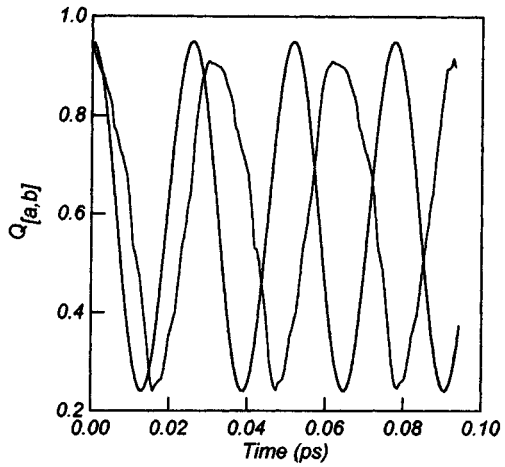

FIG. 4. Free electron charge density oscillations (black line) compared with electron-in-exciton charge density oscillations (gray line) in the left well.

number of tunneling events. Second, the total density oscillation amplitude (the product of the direct created exciton concentration and the electron-in-exciton charge oscillations) can be strongly damped by the exciton-free carriers interaction. This effect has been calculated for photoexcited electrons in ACQW.[16] Thus, the number of perceptible oscillations is reduced to a few tens, in good agreement with experiments.

IV. CONCLUSIONS

We have numerically integrated in space and time the effective mass Schrödinger equation for a directly created electron-hole pair in an asymmetric double quantum well system. This exciton is generated through hole-assisted electron resonant tunneling, being a two-particle process. We have obtained the excitonic dynamics, which is basically determined by the two different natures of the exciton (spatially indirect and spatially direct exciton). The electron-in-exciton asymmetric charge oscillations, together with the confined hole, will lead to a new type of coherent electromagnetic radiation emerging from the semiconductor ACQW, after electrical pumping. Taking into account that the mean life for an electron-hole pair in GaAs is 200 ps, an experimental observation of such a process is possible in principle. Other quantities as current densities (which are directly translated into THz radiation) can be straightforwardly usable for experimentalists. A forthcoming publication will deal with this question more profusely.

ACKNOWLEDGMENTS

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