Self-Pulse-Shaping Coherent Control of Excitons in a Semiconductor Microcavity

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The coherent control of the exciton population in quantum wells (QW's) is considered. For cavity-free QW's a 0π pulse leaves the QW empty of excitons subsequent to its passage. For the microcavity, in addition to certain 0π pulses, specially tailored pulses with nonzero area are also optimal. The coherent depopulation is a strong-coupling effect and is due to the π phase shift in the self-consistent Maxwell field (i.e., including the local-field effect associated with the exciton resonance) in the cavity that occurs in the course of a vacuum-field Rabi flop. [S0031-9007(96)01755-3]

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Recently, Heberle *et al.* demonstrated the use of phase-controlled optical pulse pairs first to populate coherently and then to depopulate a quantum well (QW) with excitons—electron-hole pairs bound by the Coulomb interaction—on a 100-fs time scale [1]. In these experiments, one utilizes a pair of time-delayed identical pulses but with various relative phase shifts. When the phase shift is π, as shown in Fig. 1(a), the first pulse generates excitons, and the second pulse coherently depopulates the QW. This effect is understandable within linear optics; the two pulses create in the medium polarizations of equal amplitude but with a π phase shift, resulting in destructive interference and thus coherent depopulation of the QW. In fact, a π-shifted pulse pair is only a specific member of a more general class of pulses that results in this effect. The background is given in work on the propagation of 0π optical pulses in atomic vapors [2,3]. It was shown that a so-called 0π pulse can be formed, in which the first half of the pulse is absorbed by the atoms, and the second π-shifted half returns the atoms to the ground state.

In the past few years such concepts have been applied to GaAs/AlGaAs multiple QW's [1,4]. The use of phase-controlled pulses to control the exciton population in QW's—and thus optical nonlinearities—has been proposed as the basis for high-speed all-optical switching [1]. We have recently shown, however, that severe constraints imposed by dephasing and the optical nonlinearity in GaAs/AlGaAs QW's may prohibit practical application of standard QW's in room-temperature 100-gigabit/sec optical communications systems [5]. By embedding the QW within a resonant planar optical microcavity, though, the strong enhancement of the local field at the QW leads to an increased resonant nonlinear optical cross section with respect to the externally applied field [6]. Thus a marriage of semiconductor microcavities and coherent control is promising for the next generation of high-speed optical switches. To utilize microcavities in this application, however, we need pulse shapes, which, like 0π pulses in cavity-free media, result in the system being returned to the ground state following the passage of the pulse. In addition, coherent control offers a means to measure dephasing rates where other techniques, such as four-wave mixing, may be complicated by the presence of the cavity [1].

In this Letter, we identify for the first time a pulse shape of *nonzero* area that results in the coherent deexcitation of the cavity-embedded QW. For comparison, we discuss the coherent exciton population dynamics in a QW in the presence of a sub-ps optical pulse in two situations: a QW without any additional structure for electromagnetic (EM) confinement and a QW embedded within an optical microcavity. We first show for the cavity-free case the

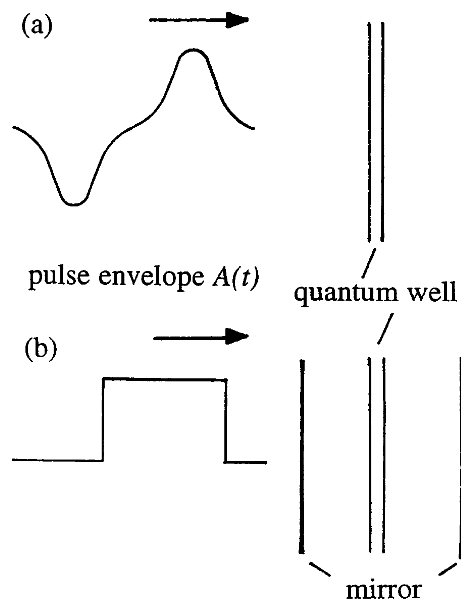

FIG. 1. Schematic of the use of phase-controlled pulses to coherently populate and depopulate a QW of excitons. For the cavity-free case (a) a pair of π-phase-shifted pulses (special case of 0π pulse) is used. The first pulse populates; the second depopulates the QW. For the cavity-embedded QW (b) an externally applied 0π pulse does not result in the coherent depopulation of the QW since the Maxwell field at the QW location in the cavity is not a 0π pulse. Instead, a square pulse whose duration is an integer number of VFR flops results in a 0π Maxwell field at the QW location.

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0π pulse is indeed optimal to depopulate the QW. For the microcavity, in addition to certain 0π pulses, *other pulse shapes with nonvanishing area are also found to be optimal.* In particular, a resonant pulse with a square envelope, as shown in Fig. 1(b), with a duration given by the time to undergo an integer number of vacuum-field Rabi (VFR) flops [7] leaves the QW empty of excitons. Note that the coherent depopulation in this case is achieved in the weak-field limit with a pulse of nonzero area. *This effect occurs because for the square pulse the self-consistent Maxwell field in the cavity is in fact a 0π pulse even though the externally applied pulse is not.* To occur, this self-pulse shaping requires both the cavity and the QW to be present. In other words, the local-field effect associated with the presence of the exciton resonance is an essential feature.

There has been a tremendous growth in activity and interest in the past few years in semiconductor microcavities spurred in part by the prospect of observing quantum-optical effects, such as have been predicted for atom-cavity systems, but also by the unique interplay of solid-state effects with strongly confined photons. Semiconductor microcavities [8] consist of one or several QW's monolithically buried between distributed Bragg reflectors to form a planar optical cavity of width on the order λ. If the cavity EM resonance at normal incidence is degenerate with the exciton, the reflection spectrum displays two dips equally spaced about the degenerate exciton-cavity resonance and split by the VFR splitting $2\hbar\Omega$ [8]. These two dips are direct evidence of the normal modes formed by the coupled exciton-cavity system, and are known as cavity polaritons (CP) [9]. Because the physics of the CP modes may be described in terms of coupled harmonic oscillators, it is equally valid to think of these modes as the elementary excitations of the system or as the EM resonances of the exciton-cavity system. We shall find the latter viewpoint more convenient.

To model the coherent control of the exciton population, we employ the coupled Maxwell and semiconductor Bloch equations (SBE) in the low-density limit projecting out only the lowest-lying exciton. The first task is to obtain in linear optics the dynamical dipolar response associated with the exciton–crystal-ground-state transition for a monochromatic EM field. We then obtain the induced polarization, which determines the exciton population via the SBE. We consider normally incident EM fields and assume a single QW of thickness $L_z \ll \lambda\!\!\!^{-} = \hbar c/(E_{\rm ex} n_b)$ ($\geq$400 Å) [10] in the material corresponding to the exciton–crystal-ground-state transition with $E_{\rm ex}$ the transition energy, c the speed of light, and n_b the background refractive index.

From the theory of the nonlocal optical response of a QW, the polarization density $P(\varepsilon, z)$ induced by an incident electric-field amplitude $E_{\rm inc}(\varepsilon, z')$ oscillating at frequency $\varepsilon/\hbar$ is [11]

$$P(\varepsilon, z) = g e^2 \mu^2 |F_{\rm ex}(\mathbf{0})|^2 f_c^*(z) f_v(z) \times \left[\int_{-\infty}^{\infty} dz' f_c(z') f_v^*(z') (\varepsilon - E_{\rm ex} + i\Gamma_{\rm sc})^{-1} \times E_{\rm tot}(\varepsilon, z') \right], \quad (1)$$

where g is a spin-orbit factor (=1 for heavy-hole excitons), $e\mu$ is the dipole matrix element between the s-like conduction- and p-like bulk valence-band Bloch states, $F_{\rm ex}(\mathbf{r}_\parallel)$ is the exciton envelope function, f_c (f_v) is the envelope function for the conduction (valence) subband of interest, and $\Gamma_{\rm sc}$ is a phenomenological nonradiative damping. The dephasing time is related to $\Gamma_{\rm sc}$ by $2\Gamma_{\rm sc}/\hbar = T_2^{-1}$. $\Gamma_{\rm sc}$ accounts for scattering which either directly destroys the phase of the excitation or scatters the state to a wave vector $\mathbf{k} \neq 0$. Dephased excitons contribute to the incoherent part $N_{\rm incoh}$ of the population N and thus these excitons cannot be coherently depopulated. $E_{\rm tot}(\varepsilon, z')$ is the dependence in the direction normal to the QW plane of the total (i.e., Maxwell) electric field. The areal polarization density is $P(\varepsilon) = \int_{-\infty}^{\infty} dz\, P(\varepsilon, z)$. Since $L_z \ll \lambda\!\!\!^{-}$, $E_{\rm tot}(\varepsilon, z')$ is effectively constant across the QW. From Eq. (1) we obtain

$$P(\varepsilon) = g e^2 \mu^2 |F_{\rm ex}(\mathbf{0})|^2 |S|^2 (\varepsilon - E_{\rm ex} + i\Gamma_{\rm sc})^{-1} \times E_{\rm tot}(\varepsilon, z_{\rm QW}), \quad (2)$$

where $z_{\rm QW}$ is the position of the QW and $S = \int dz\, f_c^*(z) f_v(z) \approx 1$.

In the cavity-free case, Eq. (2) together with the continuity of $E_{\rm tot}$ gives $E_{\rm tot}(\varepsilon, z_{\rm QW}) = t_{\rm QW}(\varepsilon) E_{\rm inc}(z_{\rm QW})$ with $t_{\rm QW}(\varepsilon) = (\varepsilon - E_{\rm ex} + i\Gamma_{\rm sc})/(\varepsilon - E_{\rm ex} + i\Gamma)$ the electric-field transmission coefficient [12], $\Gamma = \Gamma_{\rm sc} + \Gamma_{\rm rad}$, and $\Gamma_{\rm rad} = 2\pi g e^2 \mu^2 E_{\rm ex} |F_{\rm ex}(\mathbf{0})|^2 |S|^2/(\hbar c n_b)$ the radiative width of $\mathbf{k} = 0$ excitons obtained from the theory of QW exciton polaritons [13]. This gives $P(\varepsilon) = \chi(\varepsilon) E_{\rm inc}(z_{\rm QW})$ with $\chi(\varepsilon) = g e^2 \mu^2 |F_{\rm ex}(\mathbf{0})|^2 |S|^2/(\varepsilon - E_{\rm ex} + i\Gamma)$ the susceptibility relating $P(\varepsilon)$ and $E_{\rm inc}(z_{\rm QW})$. In the time domain,

$$P(t) = \int_{-\infty}^{\infty} dt'\, \chi(t') \mathcal{E}_{\rm inc}(t - t'). \quad (3)$$

Here $\mathcal{E}_{\rm inc}(t) = A(t) \exp(-i\omega_0 t)$ is the incident electric field at the location $z_{\rm QW}$ of the QW, $A(t)$ is the pulse envelope, and ω_0 is the carrier frequency. $\chi(t)$ is just the time-Fourier transform of $\chi(\varepsilon)$. Explicitly, we have

$$\chi(t) = -i g e^2 \mu^2 |F_{\rm ex}(\mathbf{0})|^2 |S|^2 e^{-i(E_{\rm ex} - i\Gamma)t/\hbar} \theta(t), \quad (4)$$

where $\theta(t)$ is the Heaviside step function.

The SBE imply that the coherent part $N_{\rm coh}(t)$ of the population is proportional to $|P(t)|^2$ [14], while the total population $N(t) = N_{\rm coh}(t) + N_{\rm incoh}(t)$, where the incoherent part $N_{\rm incoh}(t)$ consists of excitons that have dephased. Only the coherent part interferes with the optical field although the *total* population $N(t)$ modulates the nonlinear optical properties [15], thus providing a probe on the dynamics [1]. Thus $N_{\rm coh}(t)$ is governed by the optical pulse, and it is here we can attain control of the population. As nonradiative dephasing occurs, $N_{\rm incoh}(t)$ is fed at the expense of $N_{\rm coh}(t)$. In order to depopulate the QW effectively, we must choose a temporal profile for $A(t)$ which minimizes $N(t)$. If the pulse duration $\tau \ll \hbar/(2\Gamma_{\rm sc})$ ($\approx$7 ps for very high quality QW's at low T) [16], then we can neglect $N_{\rm incoh}$.

Let us see what temporal profiles $A(t)$ will yield the desired result, namely, that for $t > \tau$, $N_{\rm coh}(t)$ is a minimum. Take, for example, $\omega_0 = E_{\rm ex}/\hbar$. If $t > \tau$, then Eq. (3) gives

$$P(t) = ige^2\mu^2|F_{\rm ex}(\mathbf{0})|^2|S|^2 e^{-i(E_{\rm ex}-\Gamma)t/\hbar} \times \int_{-\infty}^{\infty} dt'\, A(t') e^{\Gamma t'/\hbar}. \tag{5}$$

Since we are interested in the regime $\tau \ll \hbar/(2\Gamma_{\rm sc})$, we neglect the exponential in the integrand in Eq. (5). We conclude that to leave zero $N_{\rm coh}$ in the QW following the passage of the pulse, we require an envelope $A(t)$ whose temporal integral $P(t > \tau)$ vanishes. This is precisely a 0π pulse, as shown in Fig 1(a). The resulting dynamics of $N_{\rm coh}$ and $N_{\rm incoh}$ is more fully explored in Ref. [5].

We turn our attention to cavity-embedded QW's. The treatment given above is generalized to relate the incident field external to the cavity with the polarization and hence the population in the QW. To avoid lengthy numerical computations and yet retain a close connection with realistic structures, we take the following model for the cavity. We consider lossless planar mirrors characterized by amplitude transmission and reflection coefficients T and R which are assumed frequency independent. In addition, we assume that the QW lies centered in the cavity and that the cavity width is L. The linear pulse propagation through the cavity is modeled by a transfer matrix [12]. For an incident field of unit amplitude, the reflected amplitude R_c and transmitted amplitude T_c are given by

$$\begin{bmatrix} T_c \\ 0 \end{bmatrix} = \begin{bmatrix} \frac{T^2-R^2}{T} & \frac{R}{T} \\ -\frac{R}{T} & \frac{1}{T} \end{bmatrix} \begin{bmatrix} e^{i\kappa L/2} & 0 \\ 0 & e^{-i\kappa L/2} \end{bmatrix} \times \begin{bmatrix} 1+\chi' & \chi' \\ -\chi' & 1-\chi' \end{bmatrix} \begin{bmatrix} e^{i\kappa L/2} & 0 \\ 0 & e^{-i\kappa L/2} \end{bmatrix} \times \begin{bmatrix} \frac{T^2-R^2}{T} & \frac{R}{T} \\ -\frac{R}{T} & \frac{1}{T} \end{bmatrix} \begin{bmatrix} 1 \\ R_c \end{bmatrix}, \tag{6}$$

with $\kappa = \varepsilon/(\hbar c)$ and $\chi'(\varepsilon) = -i\Gamma_{\rm rad}/(\varepsilon - E_{\rm ex} + i\Gamma_{\rm sc})$ [12]. T_c is the inverse of the 22 entry of the product of matrices in Eq. (6) [12]. To obtain $E_{\rm tot}(\varepsilon, z_{\rm QW})$ with $z_{\rm QW} = L/2$, we propagate the field on the left-hand side of Eq. (6) to the position of the QW, to give $E_{\rm tot}(\varepsilon, z_{\rm QW}) = (T_c/T)[\exp(-i\kappa L/2) + R\exp(i\kappa L/2)]$. From Eq. (6), the cavity transmission coefficient is $T_c = T^2/[(1-\chi')\exp(-i\kappa L) - R^2(1+\chi')\exp(i\kappa)L - 2R\chi']$. We consider an $L = \lambda/2$, high-Q cavity $[L = \pi\hbar c/(n_b E_{\rm ex}),\ R \to -1]$, in which the exciton and cavity modes are degenerate. We have

$$T_c = -i\frac{T^2}{2\pi}\left(\frac{E_{\rm ex}}{\varepsilon - E_{\rm ex} + i\Gamma_{\rm cav} - 2\pi^{-1}\Gamma_{\rm rad}E_{\rm ex}/(\varepsilon - E_{\rm ex} + i\Gamma_{\rm sc})}\right), \tag{7}$$

where the cavity resonance width $\Gamma_{\rm cav} = E_{\rm ex}/(2Q) = (E_{\rm ex}/\pi)(1-R^2)/(1+R^2)$.

We may now combine the previous results to get $E_{\rm tot}(\varepsilon, z_{\rm QW}) = -2iT_c/T$, and using Eq. (2), $P(\varepsilon) = -2ige^2\mu^2|F_{\rm ex}(\mathbf{0})|^2|S|^2(T_c/T)E_{\rm ex}/(\varepsilon - E_{\rm ex} + i\Gamma_{\rm sc})$. Fourier transforming to the time domain (δ-function incident pulse) gives

$$T_c(t) = (2\pi)^{-1}T^2 E_{\rm ex}\cos\Omega t e^{-i(E_{\rm ex}-i\overline{\Gamma})t/\hbar}\theta(t), \tag{8a}$$

$$\chi(t) = i(2\pi^2\hbar\Omega)^{-1}ge^2\mu^2|F_{\rm ex}(\mathbf{0})|^2|S|^2TE_{\rm ex} \times \sin\Omega t e^{-i(E_{\rm ex}-i\overline{\Gamma})t/\hbar}\theta(t), \tag{8b}$$

where we have assumed $\Gamma_{\rm cav}, \Gamma_{\rm sc} \ll \hbar\Omega$ with $2\hbar\Omega = \sqrt{8\Gamma_{\rm rad}E_{\rm ex}/\pi}$ the VFR splitting and $\overline{\Gamma} = \frac{1}{2}(\Gamma_{\rm cav} + \Gamma_{\rm sc})$ the average linewidth [17]. Equations (8a) and (8b) show that $T_c(t)$ [and thus $E_{\rm tot}(t, z_{\rm QW})$] and $\chi(t)$ all display VFR flopping under impulsive excitation. As expected, the polarization is in quadrature with $T_c(t)$ and $E_{\rm tot}(t, z_{\rm QW})$.

As in the cavity-free case, the time-domain version of Eq. (2) holds. We see, however, the pulse envelope $A(t)$ required to minimize the polarization left in the QW subsequent to the passage of the pulse is not as simple as the cavity-free case. In particular, for pulses with center frequency $\omega_0 = E_{\rm ex}/\hbar$ we have for $t > \tau$

$$P(t) = i(2\pi^2\hbar\Omega)^{-1}ge^2\mu^2|F_{\rm ex}(\mathbf{0})|^2|S|^2TE_{\rm ex}e^{-i(E_{\rm ex}-i\overline{\Gamma})t/\hbar} \times \int_{-\infty}^{\infty} dt'\sin[\Omega(t-t')]A(t')e^{\overline{\Gamma}t'/\hbar}. \tag{9}$$

Thus, it is clear that an arbitrary 0π pulse will not do to leave the QW with a vanishing population of excitons following the passage of the pulse; general pulse forms produce a ringing of the cavity at the VFR frequency [7]. If $\tau \ll \hbar/\overline{\Gamma}$, then the exponential in the integral in Eq. (9) may be dropped. In this case, the coherent depopulation of the QW instead can be achieved by a *square pulse envelope* [18], as shown in Fig. 1(b), whose duration $2\pi l/\Omega$ is an integer number l of VFR flops to ensure that the integral in Eq. (9) vanishes. *Such a pulse envelope has nonvanishing area; however, the phase change during the course of a VFR flop provides the phase shift to depopulate the QW coherently.* We can see from Eq. (8b) that if $A(t)$ is a square envelope of duration $2\pi l/\Omega$, then the Maxwell field at z_{QW} is in fact a 0π pulse. In other words, $A(t)$ acts as a temporal window to produce at z_{QW} a 0π pulse [19].

It is clearly advantageous to have $\hbar\Omega$ as large as possible so that dephasing during the incidence of the pulse may be minimized ($l = 1$). One way to increase $\hbar\Omega$ is to utilize n closely spaced QW's, in which case $\hbar\Omega$ is enhanced by a factor of $\sqrt{n}$ [8,9]. It is also noteworthy that for the cavity, large-bandwidth sub-fs pulses are useful for coherent control whereas for the cavity-free QW they are not; if the bandwidth is too high, then interband excitations other than the exciton of interest are also excited in the cavity-free QW. For the microcavity, however, a high-Q cavity acts as an effective spectral filter.

To conclude, we have presented a rigorous polariton-based theory of the coherent linear interaction of excitons and sub-ps optical pulses in cavity-free and cavity-embedded QW's. Complete information within the model is obtained on the EM and material degrees of freedom. For the cavity-free QW, it was shown accounting for propagation effects in the nonlocal medium, that 0π pulses of duration much less than the characteristic dephasing time result in effective coherent depopulation of the QW following its passage. For the cavity-embedded QW, however, we find as well an optimal pulse with a square envelope whose duration is an integer number of VFR flops. This results in a self-consistent Maxwell field at the location of the QW that is a 0π pulse, even though the incident pulse has nonzero area. This phenomenon is a strong-coupling effect. It cannot be arrived at by considering the cavity to be a spectral filter; the CP modes must be considered. This means that the self-shaping of the Maxwell field requires the presence of both the cavity and the QW to occur. Thus, by judicious choice of pulse shape, coherent control of excitons—which shows promise in high-speed optical switching applications—is feasible.

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