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# Exploiting exciton-exciton interactions in semiconductor quantum dots for quantum-information processing

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We propose an all-optical implementation of quantum-information processing in semiconductor quantum dots, where electron-hole excitations (excitons) serve as the computational degrees of freedom (qubits). We show that the strong dot confinement leads to an overall enhancement of Coulomb correlations and to a strong renormalization of the excitonic states, which can be exploited for performing conditional and unconditional qubit operations.

Coherent-carrier control in semiconductor nanostructures has recently attracted enormous interest[1–3] because it allows the coherent manipulation of carrier wave functions on a timescale shorter than typical dephasing times. Such a controlled ''wave function engineering'' is an essential prerequisite for a successful implementation of ultrafast optical switching and quantum-information processing in a pure solid state system. In this respect, semiconductor quantum dots (QD's) appear to be among the most promising candidates, since the strong quantum confinement gives rise to a discrete atomiclike density of states and, in turn, to a suppressed coupling to the solid-state environment (e.g., phonons).[4,5] It thus has been envisioned that optical excitations in QD's could be successfully exploited for quantum-information processing.[6,7]

Recently, optical spectroscopy of single QD's has revealed a surprisingly rich fine structure:[8–11] With increasing excitation density, additional emission peaks appear in the spectra, which supersede the original lines at even higher densities. It has been demonstrated that these optical nonlinearities are due to the strong quantum confinement, which results in a strongly reduced phase space and an overall enhancement of Coulomb correlations.[12,14] In fact, whenever one additional electron-hole pair (exciton) is added to the QD, the optical spectrum must change because of the resulting additional exciton-exciton interactions.

In this Rapid Communication we show that such optical nonlinearities in semiconductor QD's can be successfully exploited for performing elementary quantum-computational operations (quantum gates). More specifically, we shall identify the different excitonic states in QD's with the computational degrees of freedom (qubits), and we will show for a prototypical dot structure that a coherent manipulation of biexcitonic resonances can perform conditional two-qubit operations. Indeed, such controlled-NOT (C-NOT) operations, which form a cornerstone for any implementation of quantum-information processing, naturally arise from the strong internal interaction channels in the electron-hole system. To complete the requirements for implementing a universal set of quantum gates, we will demonstrate that single qubits can be manipulated by performing two C-NOT operations at different photon energies. Our model for calculating the correlated electron-hole states in the dot is based on realistic material and dot parameters; this will allow us to argue that an implementation of the proposed scheme should be possible with the present state-of-the-art sample growth and coherent-carrier control.

The initial ingredients of our calculations are the single-particle states, which we numerically obtain for a prototypical dot confinement that is parabolic in the $(x,y)$ plane and boxlike along $z$; such confinement potentials have been demonstrated to be a particularly good approximation for various kinds of self-assembled dots.[4,15] We compute the single-particle states $|\mu_{e,h}\rangle$ and energies $\epsilon^{e,h}_{\mu_{e,h}}$ for electrons and holes by numerically solving the three-dimensional single-particle Schrödinger equation within the envelope-function and effective-mass approximations.[16]

Let us suppose that the dot is subjected to a series of short laser pulses. We describe the light-matter coupling within the usual dipole and rotating-wave approximations:

$$\mathcal{H}_{\rm op} = -\frac{1}{2}\sum_i \mathcal{E}_o^{(i)}(t)\left(e^{i\omega^{(i)}t}\hat{P} + e^{-i\omega^{(i)}t}\hat{P}^\dagger\right), \qquad (1)$$

where $\mathcal{E}_o^{(i)}(t)$ is the time envelope of the $i$th laser pulse with central frequency $\omega^{(i)}$, and $\hat{P} = \Sigma_{\mu_e,\nu_h} M^*_{\mu_e\nu_h} d_{\nu_h} c_{\mu_e}$ is the interband-polarization operator, with $M_{\mu_e\nu_h}$ the dipole-matrix element for the optical transition between $\mu_e$ and $\nu_h$; the field operators $c^\dagger_{\mu_e}$ $(d^\dagger_{\nu_h})$ create an electron in state $\mu_e$ (hole in state $\nu_h$). To illustrate the action of $\mathcal{H}_{\rm op}$ on the quantum dot, let us assume that before the first laser pulse the dot is in its ground state $|{\rm vac}\rangle$ (i.e., no electrons in the conduction band and no holes in the valence band). On arrival of the first pulse, the laser light will create (through $\hat{P}^\dagger$) an interband polarization, i.e., a superposition between the vacuum state and the different excitonic states, which will propagate in phase with the driving laser. It is precisely this coherent evolution of the interband polarization, which then allows the subsequent pulses to coherently manipulate the correlated QD electron-hole states and to perform specific quantum-computational operations.

As an important step within our proposal, we use the fact that in most semiconductors, electron-hole pairs with given spin orientation can be selectively created by photons with a

 

well-defined circular polarization. Throughout this paper, we shall only consider excitons with parallel spin orientations because of their strongly reduced available phase space and the resulting simplified optical density of states.[12] Moreover, as will be discussed further below, within the proposed scheme we can restrict ourselves to single excitons and biexcitons. We expand the exciton and biexciton states within the subspaces of spin-selective electron-hole excitations, i.e., $\Psi_{\mu_e}^{\nu_h;x}$ and $\Psi_{\mu_e\mu_e'}^{\nu_h\nu_h';\lambda}$ (the latter states are antisymmetric with respect to the exchange of the two electron and hole coordinates). The excitonic eigenenergies and eigenstates are then obtained from the solutions of the two- and four-particle Schrödinger equations:

$$E_x\Psi_{\mu_e}^{\nu_h;x}=(\epsilon_{\mu_e}^e+\epsilon_{\nu_h}^h)\Psi_{\mu_e}^{\nu_h;x}+\langle\mu_e,\nu_h|\mathcal{H}^{eh}|\overline{\mu_e},\overline{\nu_h}\rangle\Psi_{\overline{\mu_e}}^{\overline{\nu_h};x} \tag{2a}$$

$$\begin{aligned}E_\lambda\Psi_{\mu_e\mu_e'}^{\nu_h\nu_h';\lambda}=&(\epsilon_{\mu_e}^e+\epsilon_{\mu_e'}^e+\epsilon_{\nu_h}^h+\epsilon_{\nu_h'}^h)\Psi_{\mu_e\mu_e'}^{\nu_h\nu_h';\lambda}+\langle\mu_e,\mu_e'|\mathcal{H}^{ee}|\overline{\mu_e},\overline{\mu_e'}\rangle\Psi_{\overline{\mu_e}\overline{\mu_e'}}^{\nu_h\nu_h';\lambda}+\langle\nu_h,\nu_h'|\mathcal{H}^{hh}|\overline{\nu_h},\overline{\nu_h'}\rangle\Psi_{\mu_e\mu_e'}^{\overline{\nu_h}\overline{\nu_h'};\lambda}\\&+\langle\mu_e,\nu_h|\mathcal{H}^{eh}|\overline{\mu_e},\overline{\nu_h}\rangle\Psi_{\overline{\mu_e}\mu_e'}^{\overline{\nu_h}\nu_h';\lambda}+\langle\mu_e,\nu_h'|\mathcal{H}^{eh}|\overline{\mu_e},\overline{\nu_h'}\rangle\Psi_{\mu_e\overline{\mu_e'}}^{\nu_h\overline{\nu_h'};\lambda}+\langle\mu_e',\nu_h|\mathcal{H}^{eh}|\overline{\mu_e'},\overline{\nu_h}\rangle\Psi_{\mu_e\overline{\mu_e'}}^{\overline{\nu_h}\nu_h';\lambda}\\&+\langle\mu_e',\nu_h'|\mathcal{H}^{eh}|\overline{\mu_e'},\overline{\nu_h'}\rangle\Psi_{\mu_e\mu_e'}^{\nu_h\nu_h';\lambda},\end{aligned} \tag{2b}$$

with the Coulomb matrix elements accounting for electron-electron ($\mathcal{H}^{ee}$), hole-hole ($\mathcal{H}^{hh}$), and electron-hole ($\mathcal{H}^{eh}$) interactions; in Eq. (2) an implicit summation over $\overline{\mu}$ and $\overline{\nu}$ has been assumed. In our computational approach, we keep for electrons and holes, respectively, the 10 energetically lowest single-particle states, and solve Eqs. (2a) and (2b) by direct diagonalization of the Hamiltonian matrix; this approach was recently proven successful in giving a realistic description of experiment.[13,17]

Figure 1(a) shows the linear absorption spectrum of the dot[16] as computed from the solutions of Eq. (2a).[18] We observe two pronounced absorption peaks ($X_0$ and $X_1$) with an energy splitting of the order of the confinement energy. A closer inspection of the exciton wave functions $\Psi_{\mu_e}^{\nu_h;x}$ reveals that: the dominant contribution of the ground state exciton $X_0$ is from the energetically lowest electron and hole single-particle states (with *s*-type character); the dominant contribution of the $X_1$ exciton is from the first excited electron and hole single-particle states (with *p*-type character).[12,14,19]

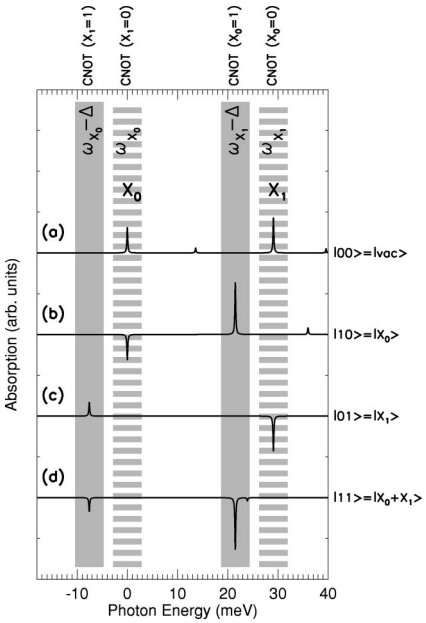


FIG. 1. Absorption spectra for semiconductor quantum dots described in the text, which are initially prepared in: (a) vacuum state; (b) exciton $|X_0\rangle$ state (exciton ground state); (c) exciton $|X_1\rangle$ state; (d) biexciton $|X_0+X_1\rangle$ state. Photon energy zero is given by the ground state exciton $X_0$ (Ref. 18).

Figure 1(b) shows the absorption spectrum for the dot initially prepared in the exciton ground state $|X_0\rangle$. Because of state filling, the character of the corresponding optical transition changes from absorption to gain (i.e., negative absorption). Moreover, the higher-energetic transition is shifted to lower energy; this pronounced redshift is attributed to the formation of a biexcitonic state, whose energy is reduced by an amount of $\Delta\approx 8$ meV because of exchange interactions between the two electrons and holes, respectively.[14] A closer inspection of the biexcitonic wave function shows that the dominant contribution is from excitons $X_0$ and $X_1$, and we therefore label this state with $|X_0+X_1\rangle$. More specifically, with the exciton creation operators $I_{X_0}$ and $I_{X_1}$ for excitons $X_0$ and $X_1$, respectively, we find that $I_{X_0}+I_{X_1}|\rangle$ has more than 95% overlap with state $X_0+X_1|\rangle$ thus providing the product-type Hibert space required for quantum-information processing. Finally, Figs. 1(c) and 1(d) report the absorption spectra for the dot initially prepared in state $X_1$ and $X_0+X_1$, respectively. We checked that optical excitations

TABLE I. Assignment of the excitonic states to the computational degrees of freedom (qubits).

| Excitonic state | Energy | Qubit state |
|---|---|---|
| $\|\text{vac}\rangle$ | 0 | $\|00\rangle$ |
| $\|X_0\rangle$ | $E_{X_0}$ | $\|10\rangle$ |
| $\|X_1\rangle$ | $E_{X_1}$ | $\|01\rangle$ |
| $\|X_0+X_1\rangle$ | $E_{X_0+X_1}=E_{X_0}+E_{X_1}-\Delta$ | $\|11\rangle$ |

within the energy region shown in Fig. 1 cannot excite any higher-excitonic states.

As the central step within the present proposal, we next assign according to Table I the excitonic states to the computational degrees of freedom (qubits). In the following, we shall demonstrate that for this choice it is indeed possible to perform the conditional and unconditional qubit operations required for quantum-information processing. We first observe in Fig. 1 that the appearance and disappearance of peaks at the frequencies indicated by the (solid and dashed) shaded areas *conditionally* depends on the setting of specific qubits: E.g., the optical transitions at $\omega_{X_0}-\Delta$ is *only* present if the second qubit is set equal to one [Figs. 1(c) and 1(d)], whereas the transition at $\omega_{X_0}$ *only* appears if the second qubit is set equal to zero [Figs. 1(a) and 1(b)]; an analogous behavior can be found for the other two transitions. Indeed, it is this conditional on and off switching of optical transitions that enables $\pi/2$ laser pulses to modify the state of one qubit or not, depending on the setting of the other qubit.

To illustrate the essence of our scheme, in the following we consider the somewhat simplified excitation scenario of a laser pulse with rectangular-shaped envelope $\mathcal{E}_o(t)$ and with central frequency $\omega_{X_0}-\Delta$, and we assume that only transitions at $\omega_{X_0}-\Delta$ are affected by the laser (results of our more realistic simulations will be presented further below). Then, the effective qubit-light Hamiltonian of Eq. (1) reads:

$$\mathcal{H}_{\rm op}=-\frac{1}{2}M\mathcal{E}_o(t)(e^{i\omega t}|11\rangle\langle 01|+e^{-i\omega t}|01\rangle\langle 11|)_{\omega=\omega_{X_0}-\Delta}, \tag{3}$$

with the dipole-matrix element $M=\langle X_0|\hat{P}|X_0+X_1\rangle$. If the duration of the pulse is $T$, its action on the system can be expressed through the unitary transformation:

$$\mathcal{U}(t>T,0)=\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & e^{-iE_{X_0}t} & 0 & 0 \\ 0 & 0 & e^{-iE_{X_1}t}\cos(\Omega_R T) & -ie^{-iE_{X_1}t}\sin(\Omega_R T) \\ 0 & 0 & -ie^{-iE_{X_0+X_1}t}\sin(\Omega_R T) & e^{-iE_{X_0+X_1}t}\cos(\Omega_R T) \end{pmatrix}, \tag{4}$$

which is supposed to act on a state vector with components $(|00\rangle,|10\rangle,|01\rangle,|11\rangle)$; here $\Omega_R=M\mathcal{E}_o$ is the Rabi frequency. Apparently, $\mathcal{U}(t,0)$ manipulates the first qubit *only* if the second qubit is set equal to one. The details of such manipulations depend on the specific choices for $t$ and $T$; e.g., for $\Omega_R T \bmod 2\pi=\pm(\pi/2)$ (i.e., $\pi/2$ pulse), $E_{X_0}t \bmod 2\pi=0$, $E_{X_1}t \bmod 2\pi=\pm(\pi/2)$, and $E_{X_0+X_1}t \bmod 2\pi=\pm(\pi/2)$, the unitary transformation of Eq. (4) precisely corresponds to a controlled NOT (C-NOT) operation.[20] By inspection of Fig. 1, we observe that a completely analogous scheme, although with different conditional qubit settings, applies for the optical transitions at different photon energies (the conditional dependence is indicated at the top of Fig. 1). Within our proposed scheme, the unconditioned qubit operations (i.e., rotation of a single qubit), which are requested besides the C-NOT operations for an implementation of a universal set of gates, can be simply achieved by combining two conditional operations at different photon energies: E.g., in order to perform a NOT operation on the first qubit (independent of the setting of the second qubit) we first have to perform a C-NOT operation at frequency $\omega_{X_0}-\Delta$, followed by a C-NOT operation at $\omega_{X_0}$ (see also Fig. 2; other single-qubit rotations can be performed by choosing different durations $T$ of the laserpulses); in principle, it is not even necessary to perform the two operations in sequence, but one could use a single appropriately tuned two-color laserpulse instead.

We finally address the possibility of experimental realization of our proposed scheme. Quite generally, the feasibility of quantum-information processing depends on the number of qubit operations which can be performed on the time scale

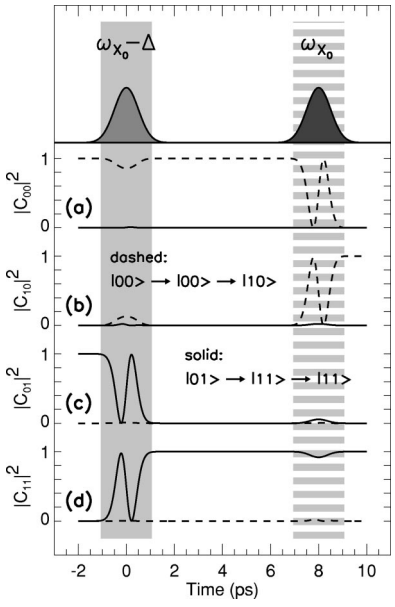


FIG. 2. Results of our simulations (neglecting dephasing) of qubit manipulations by means of coherent-carrier control. Here, $c_{00}$, $c_{10}$, $c_{01}$, and $c_{11}$ are the coefficients of the state vector corresponding to the different qubit configurations. The solid (dashed) lines present results of simulations where the system is initially prepared in the $|01\rangle$ ($|00\rangle$) state. The sequence of pulses and the corresponding photon energies are indicated at the top of the figure; for the envelopes of the laser pulses we use Gaussians $\propto\exp(-t^2/2\tau^2)$, with $\tau=0.5$ ps. The first operation at time zero corresponds to a C-NOT operation where the second qubit acts as the control qubit; the sequence of the two pulses corresponds to a NOT operation on the first qubit.

of dephasing. We therefore have to estimate the time required for performing single qubit manipulations (i.e., the duration $T_{\text{pulse}}$ of the laser pulse) and the typical dephasing times ($T_{\text{dephasing}}$). In order to perform within our scheme qubit manipulations at certain frequencies without affecting the other transitions, it is necessary that the spectral width of the laser pulse is narrow as compared to the energy separation between neighbor peaks; from the basic relation $\Delta \times T_{\text{pulse}} \sim \hbar$, we can estimate a lower limit of $T_{\text{pulse}} \gtrsim 0.1$ ps (note that much longer pulses would be required in case of non-spin-selective excitations, where the typical energy splitting of peaks in the optical spectra would be of the order of 1 meV). We also performed for the studied dot structure simulations of qubit manipulations (C-NOT and unconditioned qubit rotations). Figure 2 shows typical results of our simulations; for conceptual clarity, we have chosen rather strong laser pulses to demonstrate that even for such high electric fields the laser only manipulates components of the state vector with appropriate energies. From our simulations we safely conclude that manipulation times $T_{\text{pulse}} \approx 0.25$ ps completely suffice to suppress any population of components of the state vector at nonmatching energies. For the typical dephasing times of optical excitations in semiconductor QD's, we assume a conservative estimate of $T_{\text{dephasing}} \approx 40$ ps, which is based on the results of a recent ingenious experiment of Bonadeo *et al.*;[2] thus, $T_{\text{pulse}} \ll T_{\text{dephasing}}$. Since the primary requirement for a successful implementation of the proposed scheme is the coherent optical manipulation of single excitons and biexcitons, we strongly believe that an experimental realization is possible within the superb standards of present day coherent-carrier control. We finally stress that one of the major advantages of our proposed scheme is the fact that quantum gates can be solely performed by means of ultrashort laser pulses, and no additional external fields, which would introduce longer manipulation times and additional decoherence, are required.

In conclusion, we have presented an all-optical implementation of quantum-information processing in semiconductor QD's. Identifying the qubits with excitonic states, we have demonstrated that both conditional and unconditional qubit operations can be performed. We have discussed that the implementation of the proposed scheme is experimentally possible with the available tools of sample growth and coherent-carrier control. Generalization of our scheme to three or four qubits (i.e., three or four excitons) is possible, where further scaling is not easy because of the more complicated optical density-of-states and the resulting difficulties to make frequencies commensurable. In this respect, sample quality and the question of how many excitons can be hosted in a single dot without suffering losses due to Auger-type processes will be of essential importance. Future work will also address excitonic states in arrays of coupled semiconductor QD's, where similar excitonic renormalization effects would allow the implementation of quantum-information processing for a moderate number of qubits.

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[18] The optical absorption spectra are calculated in linear response from the imaginary part of the Fourier-transformed total interband polarization (for details see, e.g., Refs. 17,4). For conceptual clarity, in Fig. 2 we have introduced a rather large broadening of $\sim 0.2$ meV accounting for the finite lifetime of the excitonic states through environment coupling (e.g., phonons).

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[20] Note that phase changes due to the unperturbed Hamiltonian have to be taken into account. Similar problems are addressed in NMR-type implementations of quantum information processing. See, e.g., A. Steane, Rep. Prog. Phys. **61**, 117 (1998). Note that in a scenario where the superposition of states is manipulated and exploited, as requested by quantum algorithms, the phase changes due to the unperturbed Hamiltonian play an important role and unavoidably have to be taken into account. In this respect, external electric or magnetic fields could be used for a fine tuning of frequencies. It is worth mentioning that the problem of keeping track of the unperturbed time evolution is even more cumbersome in the case of NMR schemes, where the unperturbed Hamiltonian also contains terms that couple the different qubits; see, e.g., A. Steane, Rep. Prog. Phys. **61**, 117 (1998).