# Coherent population transfer in coupled semiconductor quantum dots

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We propose a solid-state implementation of stimulated Raman adiabatic passage in two coupled semiconductor quantum dots. Proper combination of two pulsed laser fields allows the coherent carrier transfer between the two nanostructures without suffering significant losses due to environment coupling. By use of a general solution scheme for the carrier states in the double-dot structure, we identify the pertinent dot and laser parameters. © *2000 American Institute of Physics.* [S0003-6951(00)05338-9]

Quantum coherence among charge states is known to have strong impact on the optical properties of confined systems. A striking example of such coherence is provided by *trapped states*,[1] where two intense laser fields drive an effective three-level system into a state which is completely stable against absorption and emission from the radiation fields. This highly nonlinear coherent-population trapping is exploited in the process of stimulated Raman adiabatic passage (STIRAP)[2] to produce complete population transfer between quantum states: Coherent control of optical excitations of atoms and molecules, as well as of chemical-reaction dynamics has been demonstrated in the last years.[2,3]

Successful implementations of such schemes in semiconductor nanostructures would have a strong impetus on the development of novel optoelectronic devices. However, in contrast to atomic systems carrier lifetimes in the solid state are much shorter because of the continuous density-of-states of charge excitations and stronger environment coupling. Although the possibility of trapped states and coherent population transfer in semiconductor quantum wells has been demonstrated theoretically,[4–6] its efficiency is expected to be extremely poor as compared to atomic systems. It has, on the other hand, been noted that the situation could be much more favorable in semiconductor quantum dots (QDs), where the strong quantum confinement leads to an atomic-like density of states and to a strongly suppressed environment coupling.[5] However, in spite of the continuing progress in dot fabrication, all the available samples still suffer from the effects of inhomogeneity and dispersion in dot size, which lead to large linewidths when optical experiments are performed on large QD ensembles. A major advancement in the field has come from different types of local optical experiments, that allows the investigation of individual QDs thus avoiding inhomogeneous broadening,[7] and simple coherent-carrier control in single dots has been demonstrated recently.[8]

In this letter, we propose a solid-state implementation of STIRAP in two coupled semiconductor QDs. Using a general solution scheme for the carrier states in the QD, which was recently proven successful in giving a realistic description of experiment,[9] we identify the pertinent dot and laser 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. In its simplest form, the STIRAP scheme involves three states, where two long-lived low-energy states $|1\rangle$ and $|3\rangle$ are dipole coupled to a state $|2\rangle$ of higher energy (which can be short lived); transitions between states $|1\rangle$ and $|3\rangle$, on the other hand, are dipole forbidden. Then, by acting with two appropriately tuned laser pulses on the system, within a STIRAP process all population is transferred between states $|1\rangle$ and $|3\rangle$ *without ever populating the ''leaky'' state* $|2\rangle$.[2] This is a remarkable property, as it allows an almost complete population transfer without suffering significant losses due to environment coupling of the interconnecting level.

Figure 1 sketches the proposed implementation of STIRAP in two coupled semiconductor QDs: The two dots (with an assumed parabolic in-plane potential[10] and a double-quantum-well confinement along $z$) are embedded in the intrinsic region of a typical $p$-$i$ field-effect structure (the dotted line in Fig. 1 indicates the Fermi energy of the $p$-type region).[11] By applying an external gate voltage, the number of surplus holes in the dot structure can be precisely controlled; throughout this letter we assume that the double dot is populated by an additional hole, where further charging is prohibited because of the Coulomb blockade. If the external electric field is strong enough, the hole wave functions become localized in either of the two dots ($|R\rangle$ or $|L\rangle$), as can be inferred from the shaded areas in Fig. 1. In contrast, the electron wave function extends over both dots (see $|e\rangle$ in Fig. 1) because of the smaller electron mass and the resulting increased interdot tunneling (electron and hole masses are different for most GaAs-based dot materials,[12,13] e.g., self-assembled $In_{0.5}Ga_{0.5}As$ dots). Thus, the interconnecting state $|2\rangle$ of the STIRAP process is assigned to the charged-exciton state [approximately consisting of one electron in state $|e\rangle$ and two holes in states $|R\rangle$ and $|L\rangle$], whereas the long-lived states $|1\rangle$ and $|3\rangle$ are assigned to the two hole states.

A detailed theoretical analysis was performed for the dot structure of Fig. 1, assuming a prototype dot confinement which is parabolic in the in-plane directions[10] and which has a profile along $z$ according to Fig. 1; such confinement po-

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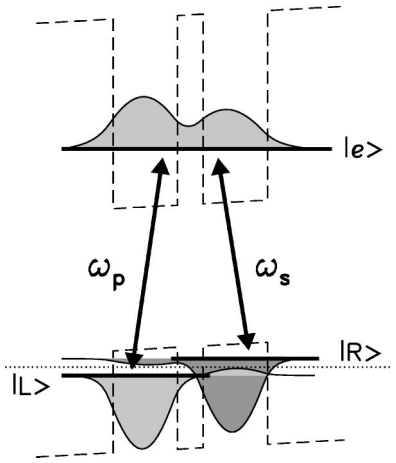


FIG. 1. Schematic illustration of the proposed double-dot structure. The dashed lines indicate the confinement potential in the $z$ direction for electron (top) and holes (bottom) including an external electric field of 1.5 mV/nm (the well widths are 5 nm and the distance between the two dots is 2 nm) (Ref. 10). The hole wave function $|R\rangle(|L\rangle)$ is localized in the right (left) dot (shaded areas; for clarity the energy splitting between the two hole states is increased here by a factor of 5); because of the lighter electron mass, the electron wave function $|e\rangle$ extends over both dots. The dotted line indicates the Fermi energy of the $p$-doped region; $\omega_s(\omega_p)$ indicates the frequency of the Stokes (pump) laser.

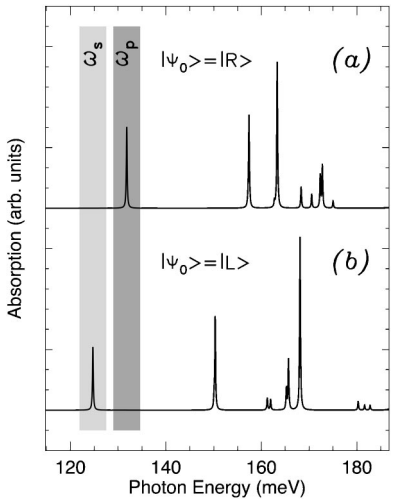


FIG. 2. Optical absorption spectrum for double-dot structure of Fig. 1 and for hole in the: (a) right-dot state $|R\rangle$; (b) left-dot state $|L\rangle$. Photon energy zero is at the semiconductor band gap; $\omega_s(\omega_p)$ indicates the photon energy of the Stokes (pump) laser. We use $\epsilon_R = 25.62$ meV, $\epsilon_L = 32.71$ meV, $\epsilon_{X^+} = 157.41$ meV for the energies (measured with respect to the semiconductor bandgap) and $M_{R,X^+} = 0.90\ \mu_o$, $M_{L,X^+} = 0.79\ \mu_o$ for the optical elements, with $\mu_o$ the dipole matrix element of the bulk material.

tentials have been demonstrated to be a good approximation for self-assembled quantum dots formed by strained-layer epitaxy.[12] In our calculations for the double dot, we start from the single-particle states derived by solving the single-particle Schrödinger equation within the envelope-function and effective-mass approximations. For electrons and holes, respectively, we keep the 20 energetically lowest single-particle states (i.e., two subbands in the $z$ direction and 10 ''Fock–Darwin'' states of the two-dimensional (2D)-harmonic oscillator[12]) and compute the charged-exciton states $|X^+\rangle$ through direct diagonalization of the Hamiltonian matrix.[9,14] Within this scheme the truncation of the Hilbert space is the *only* approximation adopted, and all Coulomb-induced effects are accounted for in a first-principles manner. For simplicity, in the following we only consider charged exciton states where the two holes have parallel spin orientations; experimentally, this can be achieved by (i) aligning the spin of the surplus hole by a small external magnetic field, and (ii) photogenerating electron–hole pairs with appropriate spin orientation by use of circularly polarized light.

Suppose that initially the surplus hole is in state $|h\rangle$ (with $h = L,R$). The linear absorption spectrum[12]

$$\alpha_h(\omega) \propto \sum_{X^+} |M_{h,X^+}|^2 \mathcal{D}_\gamma(\epsilon_h + \omega - \epsilon_{X^+}) \tag{1}$$

then provides information about the dipole–allowed transitions to charged exciton states $|X^+\rangle$; here, $M_{h,X^+}$ is the optical dipole element for the transition between states $|h\rangle$ and $|X^+\rangle$, $\epsilon_h$ and $\epsilon_{X^+}$ are the energies of the hole and the charged-exciton states, respectively, and $\mathcal{D}_\gamma(\omega) = 2\gamma/(\omega^2 + \gamma^2)$ is a broadened delta function, where the phenomenological damping constant $\gamma$ accounts for interactions with the dot environment ($\hbar = 1$). Figure 2 shows the calculated linear absorption spectra for the double dot of Fig. 1, with the surplus hole in the lowest-energy Fock–Darwin state (with $s$-type symmetry) and: (a) in the right dot; (b) in the left dot. One observes two peaks at photon energies of ~130 meV and a peak multiplet at higher photon energies. A closer inspection of the charged-exciton states reveals that: (i) in Fig. 2(a) [Fig. 2(b)] the peak at $\omega_p(\omega_s)$ corresponds to a transition from state $|R\rangle$ ($|L\rangle$) to the charged-exciton state of lowest energy; (ii) the peaks at higher photon energies correspond to the transitions to excited charged-exciton states. We thus conclude that indeed the charged-exciton ground state optically couples to both the right-dot and left-dot hole ground state. Furthermore, since all excited states are optically well separated from the two peaks at $\omega_s$ and $\omega_p$, in the following it suffices to consider as an effective three-level scheme the charged-exciton ground state only (henceforth denoted with $|X^+\rangle$), and for the hole state in the right and left dot, respectively, the Fock–Darwin state of lowest energy. In this respect two points are worth mentioning: First, our findings do not depend decisively on the chosen dot parameters, but rather reflect the basic symmetries of the dot confinement; second, because of the zero-dimensional nature of carrier states in QDs there arises no conceptual problems in choosing the Coulomb-renormalized charged-exciton state as the interconnecting state—in contrast to semiconductors of higher dimensionality, where Coulomb renormalizations can spoil a clear-cut identification of an effective three-level system.

We next turn to a discussion of the STIRAP process, where the hole is transferred from state $|R\rangle$ to state $|L\rangle$ by coupling to the intermediate charged-exciton state $|X^+\rangle$. Suppose that the system is subjected to two laser pulses, where the frequency of the ''Stokes'' (''pump'') pulse is tuned to the $L-X^+(R-X^+)$ resonance (see $\omega_s$ and $\omega_p$ in Figs. 1 and 2). To understand the essence of STIRAP, as a first preliminary step we assume that the Stokes pulse *only* affects the $L-X^+$ transition, whereas the pump pulse *only* affects the $R-X^+$ transition (results of our complete time simulations will be presented further below). Within the in-

teraction picture, the Hamiltonian of the effective three-level system then reads (neglecting environment coupling)[2]

$$\mathbf{H}=-\frac{\Omega_p(t)}{2}|X^+\rangle\langle R|-\frac{\Omega_s(t)}{2}|X^+\rangle\langle L|+\mathbf{H.c.}, \tag{2}$$

with the Rabi frequencies $\Omega_s(t)=\mathcal{E}_s(t)M_{L,X^+}$, $\Omega_p(t)=\mathcal{E}_p(t)M_{R,X^+}$, and the time envelopes $\mathcal{E}_{s,p}(t)$ of the Stokes and pump pulse, respectively. For time-independent envelopes $\mathcal{E}_{s,p}$ the Hamiltonian $\mathbf{H}$ of Eq. (2) can be diagonalized

$$\begin{aligned}&|a_0\rangle=\cos\theta|R\rangle-\sin\theta|L\rangle, \quad \omega_0=0,\\ &|a_\pm\rangle\propto|X^+\rangle\pm(\sin\theta|R\rangle+\cos\theta|L\rangle), \quad \omega_\pm=\pm\Omega_{\rm eff},\end{aligned} \tag{3}$$

with $|a_0\rangle$, $|a_\pm\rangle$ the eigenvectors and $\omega_0$, $\omega_\pm$ the eigenvalues, $\theta$ the mixing angle defined by $\tan\theta=\Omega_p/\Omega_s$, and $\Omega_{\rm eff}=\frac{1}{2}(\Omega_s^2+\Omega_p^2)^{1/2}$. $|a_0\rangle$ has the remarkable property of being decoupled from the laser fields, i.e., if the system is initially prepared in this coherent-superposition state it remains there at any time, since the contribution to the excited-state amplitude from both hole states interfere destructively.

The STIRAP process uses state $|a_0\rangle$ as a vehicle in order to transfer population between states $|R\rangle$ and $|L\rangle$. Since $|a_0\rangle$ does not involve contributions from the ''leaky'' state $|X^+\rangle$, the population can be directly channeled between the two long-lived hole states (i.e., without suffering losses from $|X^+\rangle$). Coherent population transfer is achieved by using two delayed laser pulses which overlap in time (see Fig. 3): Initially, the system is prepared in state $|R\rangle$. When the first laser pulse (''Stokes'') is smoothly turned on, the double-dot system is excited at frequency $\omega_s$. One readily observes from Fig. 2(*a*) that this pulse cannot induce any optical transition. What it does, however, is (i) to align the time-dependent state vector $|\Psi\rangle_t$ with $|a_0\rangle_t=|R\rangle$ (since $\theta=0$ in the sector $\Omega_s\neq 0$, $\Omega_p=0$), and (ii) to split the degeneracy of the eigenvalues $\omega_0,\omega_\pm$. Thus, if the second laser pulse (pump) is smoothly turned on—such that throughout $\Omega_{\rm eff}(t)$ remains large enough to avoid (nonadiabatic) transitions between $|a_0\rangle_t$ and $|a_\pm\rangle_t$—all population is transferred between states $|R\rangle$ and $|L\rangle$ within an adiabatic process where $|\Psi\rangle_t$ directly follows the time-dependent trapped state $|a_0\rangle_t$.

Figure 3 shows a time simulation of STIRAP accounting for all possible dipole transitions. Indeed, for the realistic dot and laser parameters listed in the figure captions it is possible to perform an almost complete population transfer between the two hole states, as can be inferred from the population transients in Fig. 3. We verified that moderate modifications of the dot and laser parameters do no lead to noticeable changes in the results. Note that the small population of the leaky state $|X^+\rangle$ is due to the nonadiabatic transients of the laser pulses and to the coupling to nonresonant transitions.

In conclusion, we have proposed an implementation of stimulated Raman adiabatic passage (STIRAP) in a coupled-quantum-dot structure, which is remote doped with a single surplus hole. The right- and left-dot hole states are optically connected through the charged-exciton state, where the spin orientations of electrons and holes are controlled by a small external magnetic field and by use of circularly polarized light. Proper combination of the pulsed Stokes and pump laser fields then allows a coherent population transfer between the two dot states without suffering significant losses due to environment coupling of the charged exciton state. Finally, we have discussed that the implementation of the proposed scheme is experimentally possible with the available tools of sample growth and coherent-carrier control.

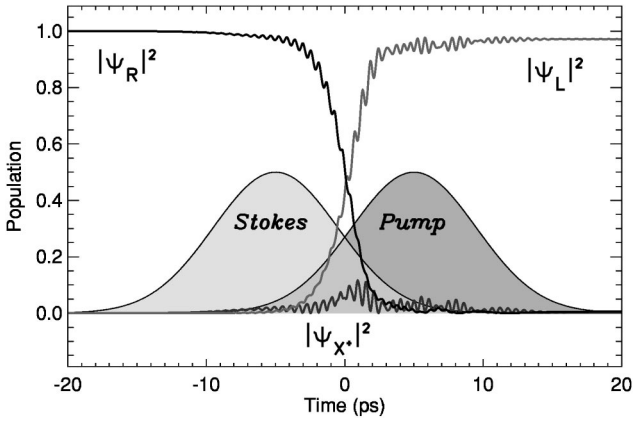


FIG. 3. Time simulation of the STIRAP process which transfers a hole from the right to the left dot, with $\Psi_h=\langle h|\Psi\rangle$ $(h=L,R)$ and $\Psi_{X^+}=\langle X^+|\Psi\rangle$. Same parameters as for Fig. 2. For the time envelopes of the Stokes and pump pulses $\mathcal{E}_{s,p}(t)$ (dashed areas) we use Gaussians centered at time $\mp 5$ ps with a full-width of half minimum of $\sim 10$ ps (with center-of-pulse Rabi frequencies $\Omega_s=\Omega_p=1$ meV), and for the effective lifetime of $X^+$ we use a relatively small value of 25 ps to account for radiative decay and other decoherence processes due to environment coupling.[8]

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[1] For a review see: E. Arimondo, Prog. Opt. **35**, 257 (1996).
[2] K. Bergmann, H. Theuer, and B. W. Shore, Rev. Mod. Phys. **70**, 1003 (1998).
[3] R. J. Gordon, Annu. Rev. Phys. Chem. **48**, 595 (1997).
[4] M. Lindberg and R. Binder, Phys. Rev. Lett. **75**, 1403 (1995); *ibid.* **81**, 1477 (1998).
[5] W. Pötz, Appl. Phys. Lett. **71**, 395 (1997); Phys. Rev. Lett. **79**, 3262 (1997).
[6] M. Artoni, G. C. La Rocca, and F. Bassani, Europhys. Lett. **49**, 445 (2000).
[7] See, e.g., K. Brunner, U. Bockelmann, G. Abstreiter, M. Walther, G. Böhm, G. Tränkle, and G. Weimann, Phys. Rev. Lett. **69**, 3216 (1992); J.-Y. Marzin, J.-M. Gérard, A. Izraël, D. Barrier, and G. Bastard, Phys. Rev. Lett. **73**, 716 (1994); Refs. in: U. Hohenester, F. Rossi, and E. Molinari, Solid State Commun. **111**, 187 (1999).
[8] N. H. Bonadeo, J. Erland, D. Gammon, D. S. Katzer, D. Park, and D. G. Steel, Science **282**, 1473 (1998); Y. Toda, T. Sugimoto, M. Nishioka, and Y. Arakawa, Appl. Phys. Lett. **76**, 3887 (2000).
[9] A. Hartmann, Y. Ducommun, E. Kapon, U. Hohenester, and E. Molinari, Phys. Rev. Lett. **84**, 5648 (2000); R. Rinaldi, S. Antonaci, M. DeVittorio, R. Cingolani, U. Hohenester, E. Molinari, H. Lipsanen, and J. Tulkki, Phys. Rev. B **62**, 1592 (2000).
[10] The confinement energies due to the in-plane parabolic potential are $\omega_0^e=30$ meV for electrons and $\omega_0^h=5$ meV for holes (with this choice, electron and hole wave functions have the same lateral extension); we use material parameters for GaAs/AlGaAs.
[11] For experimental realization of charge-tunable InGaAs quantum dots see, e.g., H. Drexler, D. Leonard, W. Hansen, J. P. Kotthaus, and P. M. Petroff, Phys. Rev. Lett. **73**, 2252 (1994).
[12] L. Jacak, P. Hawrylak, and A. Wojs, *Quantum Dots* (Springer, Berlin, 1998).
[13] D. Bimberg, M. Grundmann, and N. Ledentsov, *Quantum Dot Heterostructures* (Wiley, New York, 1998).
[14] Details of our numerical approach can be found in: U. Hohenester and E. Molinari, Phys. Status Solidi B (in press). For simplicity we do not consider electron–hole exchange interactions.