Population inversion in quantum dot ensembles via adiabatic rapid passage

E. R. Schmidgall,[1] P. R. Eastham,[2] and R. T. Phillips[1]
[1]*Cavendish Laboratory, University of Cambridge, J.J. Thomson Avenue, Cambridge CB3 0HE, United Kingdom*
[2]*School of Physics, Trinity College, Dublin 2, Ireland*
(Received 25 December 2009; revised manuscript received 11 February 2010; published 4 May 2010)

Electrical detection of adiabatic rapid passage (ARP) in quantum dot ensembles is explored by extension of a simple physical model. We include exciton and biexciton states and model the current flow into the external circuit which occurs following ionization of the exciton and tunneling of the separate carriers. The inversion produced by ARP is robust with respect to fluctuations in coupling and detuning, and we show directly that this manifests itself as only minor variation in the current drawn per dot, as the number of dots in an ensemble is increased. This approach therefore can provide a means of initializing many dots simultaneously into a specific excited state.

DOI: 10.1103/PhysRevB.81.195306 PACS number(s): 78.67.Hc, 78.20.Bh

I. INTRODUCTION

Interest in coherent manipulation of single quantum dots arises from the potential of this system to function as a qubit, the fundamental building block of a quantum computer. Experiments have demonstrated the manipulation of single quantum states through excitation with laser pulses of varying integrated pulse area.[1–3] Rabi oscillations have been observed both in the photoluminescence signal[2,3] and in the photocurrent drawn from individual quantum dots in appropriately biased structures.[1,4–7] The decrease in amplitude of Rabi oscillation with increasing pulse area observed in these experiments led to inclusion of multiple levels, and excitation-dependent effects in models of the system.[8–11] Under some circumstances, such as at higher temperatures, it has been found necessary to incorporate explicitly phonon-related effects.[12–14] It is clearly interesting to extend this approach to include manipulation of an *ensemble* of quantum dots. Such manipulation would be required, for example, to prepare an initial state in a quantum computer. It would also significantly extend our understanding of many-particle quantum dynamics, allowing the evolution of arbitrary initial states to be studied experimentally. Recent calculations (prompted by the experimental realization of quantum condensation of cavity polaritons[15–17]) have shown that if tailored occupation profiles can be created in ensembles of quantum dots a dynamical form of Bose-Einstein condensation should ensue.[18]

Previous work on coherent manipulation has focused on resonant excitation with transform-limited pulses. In this case, the excitation rotates the Bloch vector by an angle equal to the pulse area so that a π pulse inverts the dot. The same pulse, however, will not invert an off-resonant dot. Furthermore, the pulse area involves the product of the dipole moment and the integrated amplitude of the pulse. Thus the inhomogeneity in practical realizations of ensembles of dots means that no resonant pulse can invert an entire ensemble exhibiting wide variation in energy and coupling strength. There is nevertheless a very good prospect for such inversion via adiabatic rapid passage (ARP), which is known to be robust with respect to detuning and coupling variation.[19,20] In this approach, the system is driven by a chirped laser pulse whose frequency sweeps through the natural frequency of the transition to be inverted. Theoretical simulations have suggested that this technique can cause a population inversion in a quantum well[21] and control the population of systems of quantum wells.[22] Calculations have also indicated that it is possible to invert a quantum dot to the biexciton state by means of ARP.[23] In this paper, we consider the excitation of ensembles of dots by chirped pulses. We calculate the current generated in a quantum dot photodiode following such excitation, generalizing previous treatments of resonant pulses and single dots. For a single dot, we show that the robust excitonic and biexcitonic inversion expected for ARP can be observed in the photocurrent. For an ensemble of dots we show that whereas the variation in coupling and energy can suppress Rabi oscillations, the features appearing in the ARP régime remain. Thus ARP is a practical inversion process for ensembles of quantum dots, which can be monitored by electrical readout.

II. THEORETICAL MODELING

It has been shown in previous work that when linearly polarized excitation is considered, both the exciton and the biexciton levels need to be included, even at low pulse area, but other levels, such as single-particle states, are only excited to a minor extent.[8] Consequently, to model the reaction of a quantum dot ensemble to a frequency-chirped laser pulse, we consider a three-level system comprising the ground state, the exciton, and a biexciton state. The dipole coupling between these states is given by

$$\Omega_x(t) = \langle 0|\vec{\mu_x}\cdot\vec{E}(t)|x\rangle,$$

$$\Omega_b(t) = \langle x|\vec{\mu_b}\cdot\vec{E}(t)|b\rangle, \quad (1)$$

where $\vec{\mu_{(x,b)}}$ is the electric dipole moment of the exciton or biexciton transition, and $\vec{E}$ the applied electric field. Choosing the phases of the eigenstates such that $\Omega_{(x,b)}(t) = \mu_{(x,b)}E(t)$ with μ real, the Hamiltonian is

$$H = \mathcal{E}_x|x\rangle\langle x| + \mathcal{E}_b|b\rangle\langle b| - \left[\frac{E(t)}{2}(\mu_x|0\rangle\langle x| + \mu_b|x\rangle\langle b|) + \text{H.c.}\right], \tag{2}$$

where $|x\rangle$ and $|b\rangle$ are the exciton and the biexciton states, respectively, $\mathcal{E}_x$ is the energy of the exciton state and $\mathcal{E}_b$ is the energy of the biexciton state. We use the usual rotating-wave Hamiltonian so that $E(t)$ is the positive-frequency component of the complex field. For a linearly chirped Gaussian pulse, the amplitude of the electric field is given in the spectral domain by

$$E(\omega) = E_0' \exp\left[-\frac{(\omega-\omega_0)^2\tau_0^2}{2}\right]\exp\left[i\alpha'\frac{(\omega-\omega_0)^2}{2}\right]. \tag{3}$$

τ_0 is the duration of the transform-limited pulse before the chirp is applied, and α' the parameter describing the linear frequency chirp. The corresponding time domain form is[20]

$$E(t) = E_0 \exp\left(-\frac{t^2}{2\tau^2}\right)\exp(i\omega_0 t + i\alpha t^2/2), \tag{4}$$

where

$$\tau^2 = \tau_0^2\left(1 + \frac{\alpha'^2}{\tau_0^4}\right) \tag{5}$$

and

$$\alpha = \frac{\alpha'}{\tau_0^4\left(1 + \frac{\alpha'^2}{\tau_0^4}\right)} \tag{6}$$

is the temporal chirp. Note that the dimensionless temporal chirp $\alpha\tau^2 = \alpha'/\tau_0^2$ controls the dynamics [Eq. (2)]. Arbitrary values of this parameter can be achieved using linear optics with a fixed input bandwidth so only independent variations in α and τ are restricted[20] by the available bandwidth.

We transform to the interaction picture via a time-dependent unitary transformation,

$$U(t) = \exp[-i\omega(t)t|x\rangle\langle x| - 2i\omega(t)t|b\rangle\langle b|]. \tag{7}$$

Note that as usual, we have redistributed the time variation between the operators and the states; since the frequency in the transformation is the momentary frequency of the applied field, the states evolve continuously in time, and a coupling is introduced while the applied field has finite amplitude. If we set the central frequency of the chirped pulse, ω_0, resonant to the exciton transition, the Hamiltonian of the three-level system becomes

$$H_{\text{eff}} = \begin{pmatrix} 0 & -\zeta_x & 0 \\ -\zeta_x & -\hbar\alpha t & -\zeta_b \\ 0 & -\zeta_b & [\Delta - 2\hbar\alpha t] \end{pmatrix}, \tag{8}$$

where $\zeta_{(x,b)} = \frac{E_0\mu_{(x,b)}}{2}e^{-t^2/2\tau^2}$, and the biexciton detuning $\Delta = E_b - 2\hbar\omega_0$. So far we have considered an isolated system without any stochastic terms, which are needed to model dephasing, decay, and tunneling. We therefore use the standard density-matrix approach, and solve the Liouville equation,

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}[H,\rho] + L(\rho), \tag{9}$$

where ρ is the density matrix and $L(\rho)$ describes the dephasing and decay in the system. Choosing the Lindblad[24] form ensures conservation of probability, and gives

$$L(\rho) = \begin{pmatrix} 2\gamma_x\rho_{xx} & -\gamma_x\rho_{0x} & -\gamma_b\rho_{0b} \\ \cdot & (2\gamma_b\rho_{bb} - 2\gamma_x\rho_{xx}) & -(\gamma_x+\gamma_b)\rho_{xb} \\ \cdot & \cdot & -2\gamma_b\rho_{bb} \end{pmatrix} \tag{10}$$

in which we have included terms arising from transitions between levels $x \to 0$ (exciton annihilation, with rate γ_x) and levels $b \to x$ (biexciton to exciton annihilation, with rate γ_b). This form can be extended in the standard way to include terms describing pure dephasing and the two-body decay channel $b \to 0$. However, these additional processes are unlikely to be strong enough qualitatively to affect our results, and hence are excluded for simplicity. The rates γ can be split into parts which do not contribute to the photocurrent (e.g., spontaneous emission), and those which do, allowing the contribution related to tunneling of carriers out of the dot to be accounted for separately to yield a total current flow through the cycle of excitation and deexcitation.[8] The current is calculated by integrating, through the full time evolution, the rate describing tunneling out to the contacts multiplied by the relevant instantaneous density-matrix element—as detailed by Villas-Bôas *et al.*[8] Our model, with appropriate

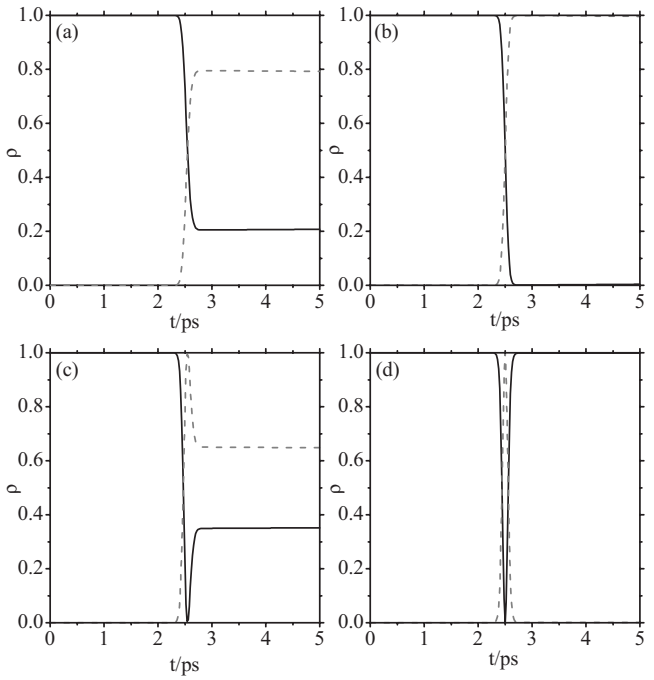

FIG. 1. The calculated diagonal density-matrix elements for the two-level model excited by a bandwidth-limited optical pulse resonant with the exciton transition, as a function of time. From (a) to (d), the optical power is increased in steps of factors of 2. The solid line represents the ground state and the dashed line the excited state.

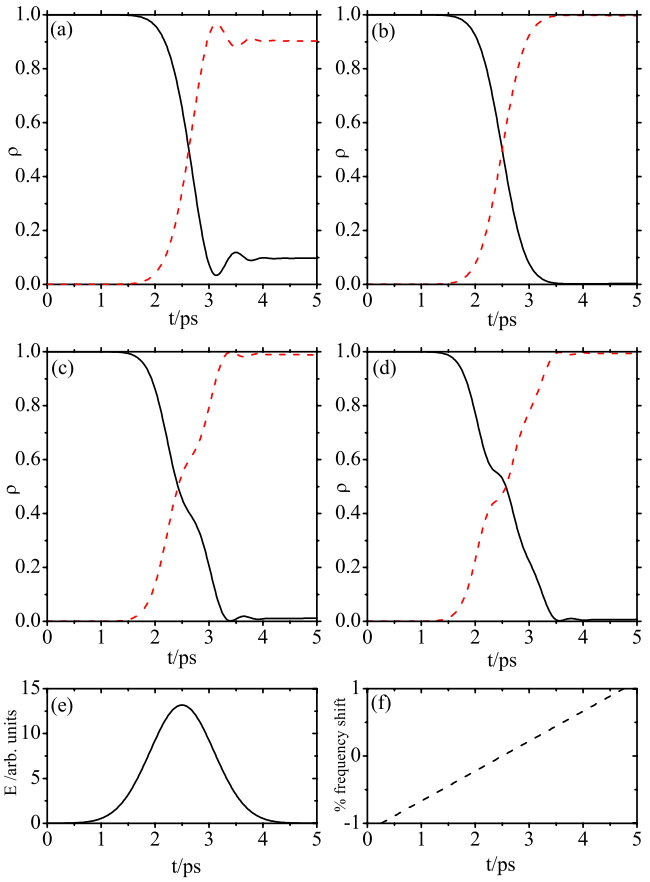

FIG. 2. (Color online) The calculated diagonal density-matrix elements for the two-level model excited by a chirped pulse as a function of time. From (a) to (d), the optical power is increased in steps of factors of 2. The solid line represents the ground state and the dashed line the excited state. (e) shows the envelope of the electric field of the chirped pulse on an arbitrary scale and (f) the linear variation in the instantaneous frequency with time through the pulse, expressed as a percentage of the central frequency.

choice of parameters, produces a description of the photocurrent comparable to that in earlier work.[8,10]

III. NUMERICAL RESULTS AND DISCUSSION

The inversion of the system in the Rabi flopping régime is sensitive to variation in both the energy and the coupling strength whereas that in the ARP régime is not.[19] We illustrate the effects of varying coupling strength in Figs. 1 and 2. In Fig. 1, the diagonal density-matrix elements for a two-level model (corresponding to a transition energy of 1450 meV) are shown for resonant excitation by a Gaussian pulse of full width at half maximum (FWHM) of 200 fs, covering four different power levels, increasing by a factor of 2 in each successive diagram. The two-level system is chosen simply because it gives a clearer picture of the inversion than the three-level model. The variation in the power can be taken to represent either a real power variation or a variation in dipole coupling as one considers different dots in an ensemble. In Fig. 1(a), the power level is set just below that required to produce the lowest-power Rabi flop. The variation in power from (a) to (d) corresponds to a factor of $\sqrt{8}$ variation in pulse area, and illustrates the fact that such variation can lead to any final state after excitation [in (b), an exciton is left; in (d), the system has returned to the ground state]. This sensitivity diminishes as the excitation passes from the normal resonant condition to that of adiabatic rapid passage. This is illustrated in Fig. 2, where once again a factor of 2 in power separates successive frames, and the pulse is derived from the 200 fs FWHM Gaussian by applying a linear chirp corresponding to $\alpha=0.05$. Here the first power chosen for the Fig. 2(a) is just below that required to

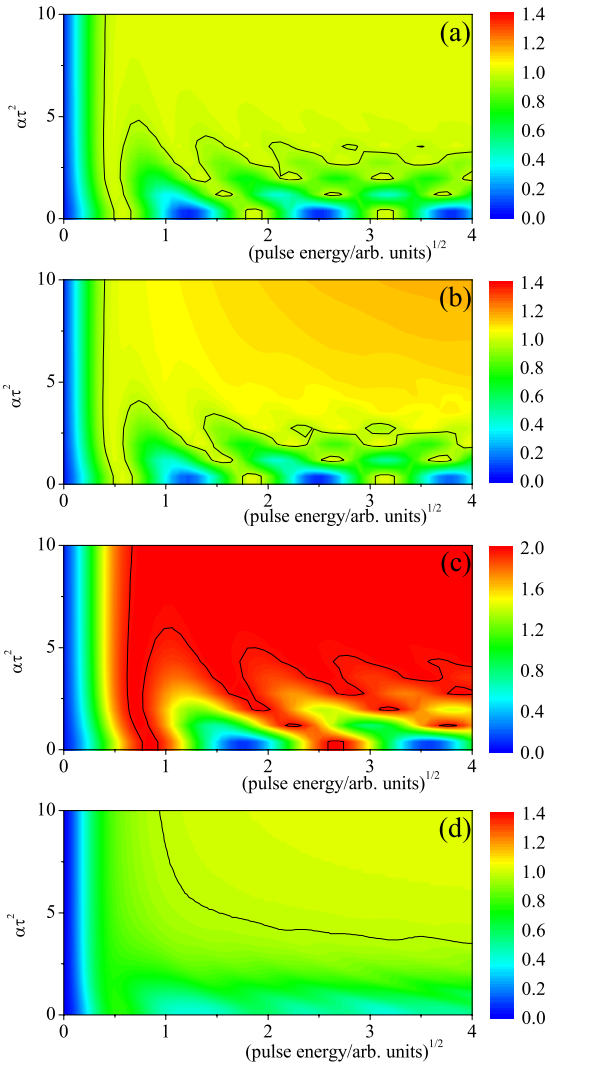

FIG. 3. (Color online) Panels (a)–(c): single-dot photocurrent as a function of the dimensionless chirp $\alpha'/\tau_0^2=\alpha\tau^2$ and pulse energy. The ratio of the exciton to biexciton coupling ζ_x/ζ_b is (a) 100, (b) 10, and (c) 1, corresponding to excitation pulses ranging from almost (a) circular to (c) linear polarization. Panel (d): photocurrent from an ensemble of 50 such dots with $\zeta_x/\zeta_b=100$. The grayscale is in electrons per dot per pulse, with a contour at 1 [for panels (a), (b), and (d)] and at 2 [for panel (c)]. In panel (d), fluctuations in the dot energies and coupling strengths preclude populating the dots with resonant pulses, destroying the Rabi oscillations. Chirped pulses however lead to robust excitonic inversion, with occupations close to one electron per dot per pulse over a large parameter region.

achieve full inversion, and increasing power by successive factors of 2 gives almost perfect inversion by ARP.

The robustness of ARP with respect to variation in the pulse power is also manifest in the calculated current drawn from a dot in a biased diode structure, shown in Fig. 3 as a grayscale map of the calculated current. In the top three panels of Fig. 3, we show the variation in photocurrent with chirp and square root of pulse power for the model of a single quantum dot. In this case, we have set the intrinsic decay time of the dot to be much longer than the time scale considered, have modeled the tunneling by a relatively weak process with time constant 100 ps, and have taken the biexciton detuning $\Delta=0$. The excitation pulse has a fixed spectral bandwidth $\tau_0=85$ fs and varying spectral chirp α'. For the top panel of Fig. 3, we have set $\zeta_x/\zeta_b=100$, in this case, the biexciton is almost completely decoupled so that along the line of zero chirp, we see the usual excitonic Rabi oscillations. Away from this region, we see large regions where the electron photocurrent is close to a value of one electron per dot per pulse, as expected when the one-exciton state is populated by ARP. Thus we see that the adiabatic population transfer is directly evident in the photocurrent; physically, this is because the tunneling time is large compared with the pulse time, and the excitation and tunneling are effectively independent. For shorter tunneling times (not shown), we find that the photocurrent can be larger than one electron per pulse because the dot can be reexcited after the first tunneling event. The effects of the biexciton on the photocurrent of a single dot are shown in panels (b) and (c) of Fig. 3, where $\zeta_x/\zeta_b=10$ and 1, respectively. In the latter case, which corresponds to excitation with linearly polarized light, there are two-photon Rabi oscillations of the biexciton along the line of zero chirp. Away from this region, the photocurrent rises to two electrons per dot, corresponding to inversion to the biexciton by ARP.[23] For intermediate polarizations [panel (b)], there is some coupling to the biexciton but it is insufficient to ensure the dynamics is adiabatic. Thus the final state with the chirped pulses is a superposition of exciton and biexciton, and the photocurrent is intermediate between the two limits. These effects are not very sensitive to the biexciton binding energy if it is within the bandwidth of the chirped pulse; clearly the biexciton will not be excited if it lies outside the pulse spectrum or if it is excluded by any other mechanism which reduces the dipole coupling. Finally we consider the case relevant to simultaneous initialization of an inhomogeneous ensemble of dots into an excited state. To illustrate this case, we again consider the three-level system but introduce 50 dots with energies taken from a Gaussian distribution of standard deviation 2 meV. This value is chosen to illustrate the effect; we obtain similar results for broader distributions provided the chirp is sufficient to span them, and the power sufficient to remain in the adiabatic régime. The exciton coupling strength is taken from a Lorentzian distribution, with a half-width one-third of the mean. (These forms of distribution are chosen as a simple approximation to the form of the result obtained from simulations of localization of excitons by interfacial roughness in narrow quantum wells but the details do not alter the result presented here.) Each biexciton coupling is taken to be 0.01 of the exciton coupling, corresponding to the single-dot case of Fig. 3(a). The result is shown in Fig. 3(d), and clearly illustrates that the inhomogeneity reduces the contrast of the Rabi fringes considerably along the line of zero chirp. However, because of the insensitivity to coupling strength and energy of the inversion by adiabatic rapid passage, the total current comes close to the expected value of 50 times the single-dot current over a wide range of chirped pulse parameters. This suggests that the photocurrent can provide a useful diagnostic of inversion in ensembles of dots provided that ARP is the means used to create inversion.

IV. CONCLUSIONS

Working from previous models for photocurrent measurement of Rabi oscillations in single-quantum dots, we have developed a model capable of treating both the case of chirped pulses and the case of quantum dot ensembles. Simulation results suggest that not only is it possible to invert single quantum dots using an adiabatic rapid-passage technique but that this technique can be applied to ensembles of quantum dots. This illustrates that by application of shaped optical pulses, ensembles of dots may be initialized as required. An extension of this idea applicable to a single dot is the use of optical excitation of constant frequency swept via the Stark effect through the optical frequency by means of a ramped electric field.

ACKNOWLEDGMENTS

This work was conducted with financial support from EPSRC under Grant No. EP/F040075/1, Science Foundation Ireland under Grant No. 09/SIRG/I1592 (P.R.E.), and the Marshall Foundation (E.S.). We thank J. Keeling for discussions.

[1] A. Zrenner, E. Beham, F. Findeis, M. Bichler, and G. Abstreiter, Nature (London) **418**, 612 (2002).
[2] T. H. Stievater, X. Li, D. G. Steel, D. Gammon, D. S. Katzer, D. Park, C. Piermarocchi, and L. J. Sham, Phys. Rev. Lett. **87**, 133603 (2001).
[3] H. Kamada, H. Gotoh, J. Temmyo, T. Takagahara, and H. Ando, Phys. Rev. Lett. **87**, 246401 (2001).
[4] E. Beham, A. Zrenner, F. Findeis, M. Bichler, and G. Abstreiter, Phys. Status Solidi B **238**, 366 (2003).
[5] E. Beham, A. Zrenner, S. Stufler, F. Findeis, M. Bichler, and G. Abstreiter, Physica E **16**, 59 (2003).
[6] Q. Q. Wang, A. Muller, P. Bianucci, E. Rossi, Q. K. Xue, T. Takagahara, C. Piermarocchi, A. H. MacDonald, and C. K. Shih, Phys. Rev. B **72**, 035306 (2005).
[7] S. Stufler, P. Ester, A. Zrenner and M. Bichler, Phys. Rev. B **72**, 121301(R) (2005).

[8] J. M. Villas-Bôas, S. E. Ulloa, and A. O. Govorov, Phys. Rev. Lett. **94**, 057404 (2005).
[9] J. M. Villas-Bôas, S. E. Ulloa, and A. O. Govorov, Physica E **26**, 337 (2005).
[10] H. S. Brandi, A. Latge, Z. Barticevic, and L. E. Oliveira, Solid State Commun. **135**, 386 (2005).
[11] D. Mogilevtsev, A. P. Nisovtsev, S. Kilin, S. B. Cavalcanti, H. S. Brandi, and L. E. Oliveira, Phys. Rev. Lett. **100**, 017401 (2008).
[12] A. Krügel, V. M. Axt, T. Kuhn, P. Machnikowski, and A. Vagov, Appl. Phys. B: Lasers Opt. **81**, 897 (2005).
[13] A. Vagov, M. D. Croitoru, V. M. Axt, T. Kuhn, and F. M. Peeters, Phys. Status Solidi B **243**, 2233 (2006).
[14] A. Vagov, M. D. Croitoru, V. M. Axt, T. Kuhn, and F. M. Peeters, Phys. Rev. Lett. **98**, 227403 (2007).
[15] R. Balili, V. Hartwell, D. Snoke, L. Pfeiffer, and K. West, Science **316**, 1007 (2007).
[16] J. Kasprzak, M. Richard, S. Kundermann, A. Baas, P. Jeambrun, J. M. Keeling, F. M. Marchetti, M. H. Szymanska, R. Andre, J. L. Staehli, V. Savona, P. B. Littlewood, B. Deveaud and L. S. Dang, Nature (London) **443**, 409 (2006).
[17] A. P. D. Love, D. N. Krizhanovskii, D. M. Whittaker, R. Bouchekioua, D. Sanvitto, S. A. Rizeiqi, R. Bradley, M. S. Skolnick, P. R. Eastham, R. André, and L. S. Dang, Phys. Rev. Lett. **101**, 067404 (2008).
[18] P. R. Eastham and R. T. Phillips, Phys. Rev. B **79**, 165303 (2009).
[19] J. S. Melinger, S. R. Gandhi, A. Hariharan, J. X. Tull, and W. S. Warren, Phys. Rev. Lett. **68**, 2000 (1992).
[20] V. S. Malinovsky and J. L. Krause, Eur. Phys. J. D **14**, 147 (2001).
[21] A. A. Batista and D. S. Citrin, Phys. Rev. B **74**, 195318 (2006).
[22] A. A. Batista, Phys. Rev. B **73**, 075305 (2006).
[23] H. Y. Hui and R. B. Liu, Phys. Rev. B **78**, 155315 (2008).
[24] G. Lindblad, Commun. Math. Phys. **48**, 119 (1976).