Rabi oscillations in a semiconductor quantum dot: Influence of local fields

G. Ya. Slepyan, A. Magyarov, and S. A. Maksimenko
Institute for Nuclear Problems, Belarus State University, Bobruiskaya 11, 220050 Minsk, Belarus

A. Hoffmann and D. Bimberg
Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstr. 36, 10623 Berlin, Germany
(Received 6 February 2004; revised manuscript received 12 April 2004; published 29 July 2004)

The influence of electron-hole dipole-dipole interactions (local fields) on the excitonic Rabi oscillations in an isolated quantum dot (QD) has been theoretically investigated. An analysis for the QD interaction with monochromatic field and ultrashort Gaussian pulse has been performed. On the basis of optical Bloch equations the Rabi oscillation dynamics has been investigated. As a result, the bifurcation and essentially anharmonic regimes in the Rabi oscillations in a QD exposed to the monochromatic field have been predicted. The strong dependence of the period of Rabi oscillations on the QD depolarization has been revealed. For the Gaussian pulse it has been shown that the final state of inversion as a function of the pulse peak strength demonstrates step-like transitions.

DOI: 10.1103/PhysRevB.70.045320 PACS number(s): 73.21.−b, 42.50.Ct, 78.67.Hc

Research on the properties of quantum dots (QDs)—nano-scale 3D confined narrow-gap insertions in a host semiconductor—has continued to grow unabated owing to the great potentiality of such structures in engineering applications, such as active media of a double heterostructure laser.[1,2] Ultrahigh material and differential gain, orders of magnitude exceeding those in quantum well lasers, has been experimentally confirmed. Recently QDs have been proposed to serve as nodes of quantum networks that store and process quantum information.[3,4] An experimental observation of a single-QD absorber has been reported in Ref. 5. The application of semiconductor QDs in cavity electrodynamics[6–8] and as potential quantum-light emitters[9,10] is now intensively discussed. Spontaneous emission in QDs[11] and electromagnetic fluctuations are also in the focus of interest. During the last decade, attempts have been made to establish the correspondence and to reveal principal differences between two-level atoms and excitons in semiconductors. Many analogies have been successfully investigated, such as the excitonic optical Stark effect, photon echoes in four-wave mixing experiments, excitonic Rabi oscillations in quantum wells[12] and QDs.[13–15] A peculiarity which, among others, distinguishes an exciton from an atomic two-level system is that the exciton exists in a medium and interacts with the medium inducing its polarization (i.e., local fields). Especially pronounced manifestation of local fields is expected in 3D confined systems, QDs. In the present paper, we investigate the role of local fields in excitonic Rabi oscillations in QDs.

As a result of the strong coupling between the incident electromagnetic field and the atomic system, electron population oscillates between excited and ground states at the Rabi frequency.[16,17] Rabi oscillations are well established in a different physical systems such as cold trapped ions,[18] Bose-Einstein condensate,[19] semiconductor quantum wells.[12] Among them, excitonic Rabi oscillations in QDs are very promising: observed experimentally in Refs. 13–15 they correspond to the one-qubit rotation that is the step towards the QD application in quantum information processing.[13] Excitonic Rabi oscillations in a QD are to be distinguished from those of an ordinary atom by the following two factors: (i) oscillator strength in a QD is essentially larger;[13] (ii) Rabi oscillation picture strongly depend on QD geometrical configuration. The first factor gives us an opportunity to observe the Rabi oscillations in the essentially smaller field, than for ordinary atoms. The second factor opens the possibility for the effect to control.

In previous investigations (see Refs. 20–26), the significant role of electron-hole dipole-dipole interaction (local field) in the electromagnetic response of different structures has been predicted. General equations for the interaction of quantum states of light with condensed matter influenced by local fields has been formulated for bulk media in Ref. 26 and for a confined QD exciton in Ref. 25. However, the application of these equations in Refs. 25, 26 were restricted to the weak light-matter coupling regime. The local field impact on the spontaneous emission decay of an excited two-level atom in the linear dielectric host has been considered in Ref. 26. In this paper it has been pointed out that the quantum theory of local fields should incorporate both quantum field theory and a many body problem. Local fields induce a fine structure of the QD absorption (emission) spectrum:[25] instead of a single line with the frequency corresponding to the exciton transition, a doublet is appeared with one component shifted to the blue (red). It has been demonstrated that in the limiting cases of classical light and single-photon states the doublet is reduced to a singlet shifted in the former case and unshifted in the latter one. Consequently, one can expect that local field effects enhanced by the strong light-QD coupling will manifest themselves in a number of observable modifications of the conventional picture of the Rabi oscillations.

To describe the strong coupling regime between the atom and the electromagnetic field, the Jaynes-Cummings (JC) model is conventionally used.[16,17] In our paper we present a microscopic theory of strong light-QD coupling restricted to the case of a classical electromagnetic field. The electron-hole dipole-dipole interactions in QD are incorporated into the JC model of excitonic Rabi oscillations. Both a QD interaction with a harmonic electromagnetic field and an ultrashort Gaussian pulse are considered. QD is modeled as a spatially confined two-level quantum oscillator. In the strong confined regime for exciton in the QD (which is considered as an electron-hole pair), the Coulomb interaction is assumed to be negligible. In this paper a 3D Cartesian coordinate system $\mathbf{u}_\alpha$ $(\alpha=x,y,z)$ is used, with the unit vector $\mathbf{u}_x$ parallel to the electron-hole pair dipole moment: $\boldsymbol{\mu}=\mu\mathbf{u}_x$. We investigate a single, arbitrary shaped QD exposed to a nonmonochromatic field linearly polarized along $\boldsymbol{\mu}$: $\mathbf{E}(t)=E_{0x}\mathbf{u}_x=\mathrm{Re}[\mathcal{E}(t)\exp(-i\omega t)]\mathbf{u}_x$, where $\mathcal{E}(t)$ is the slow-varying electric field amplitude, and ω is the field carrier frequency. According to Ref. 25, in the two-level approximation the system "QD+electromagnetic field" is described by the Hamiltonian

$$H=(\epsilon_e a_e^+ a_e+\epsilon_g a_g^+ a_g)-V\hat{P}_x E_{0x}+\Delta H, \tag{1}$$

where $\epsilon_{e,g}$ are the energy eigenvalues of the excited and ground states, respectively; $a_{g,e}^+$ and $a_{g,e}$ stand for the creation and annihilation operators of an electron in the ground and excited states; V is the QD volume; $\hat{P}_x=V^{-1}(-\mu b^+ +\mu^* b)$ is the polarization operator; $b^+=a_g a_e^+$ and $b=a_g^+ a_e$ are the creation and annihilation operators for the electron-hole pair. The first term in (1) is the Hamiltonian of the free electron-hole pair, the second term is the Hamiltonian of interaction of a pair with an electromagnetic field, while ΔH is the correction to local fields. The latter one is given by[25]

$$\Delta H=4\pi N_x P_x(-\mu b^+ +\mu^* b), \tag{2}$$

where N_x is the depolarization coefficient, and $P_x=\langle\hat{P}_x\rangle$ is the macroscopic polarization of the QD. Under the rotating wave approximation, the optical Bloch equations,

$$\frac{\partial u}{\partial t}=-\gamma_T u-\Omega_R w-\delta v-\Delta\omega(w+1)v, \tag{3}$$

$$\frac{\partial v}{\partial t}=-\gamma_T v+\delta u+\Delta\omega(w+1)u, \tag{4}$$

$$\frac{\partial w}{\partial t}=-\gamma_L(w+1)+\Omega_R u, \tag{5}$$

correspond to the Hamiltonian (1). In these equations phenomenological parameters γ_T and γ_L are the dephasing and the homogeneous broadening, respectively. Parameter $\Delta\omega=4\pi N_x|\mu|^2/\hbar V$ (Ref. 22) is the frequency shift resulting from the local field influence. Detuning $\delta=\omega-\omega_0-\Delta\omega$ is defined corrected to the shift; ω_0 is the excitonic transition frequency. Parameter $\Omega_R(t)=\mu\mathcal{E}(t)/\hbar$ is the Rabi frequency, w is the inversion, i.e., the difference between the excitonic population in the excited and the ground states; u and v are the real and imaginary parts of the nondiagonal element of the density matrix. Note that equations obtained contain nonlinear terms [the last terms in (3) and (4)]. These terms may result in appearing of nontrivial effects, such as the higher harmonics generation of the Rabi frequency or bifurcation in the oscillatory regime.

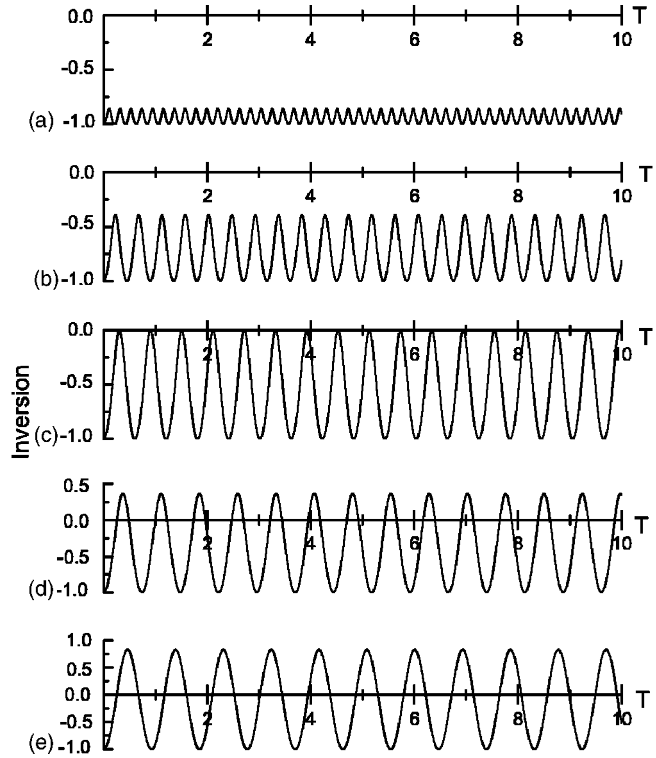

FIG. 1. Rabi oscillations of the inversion, $\delta=0$: (a) $\xi=0.002$; (b) $\xi=0.2$; (c) $\xi=0.5$; (d) $\xi=1$; (e) $\xi=3$.

We assume that the QD is in the ground state at $t=0$. Then, initial conditions for Eqs. (3)–(5) are given by

$$u(0)=v(0)=0,\quad w(0)=-1. \tag{6}$$

We start with the case of the undamped system exposed to the monochromatic field at $\delta=0$. It should be noted that the latter condition does not define the exact synchronism regime. Indeed, in the strong light-QD coupling regime the local fields lead to the substitution $\omega-\omega_0\to\delta+\Delta\omega(w+1)$ in Eqs. (3)–(5). Therefore, the actual physical detuning is not constant in time but oscillates together with the inversion.

In the presence of local fields, the analytical solution for the Bloch equations does not exist. Therefore, we have performed the numerical integration of Eqs. (3)–(5) with initial conditions (6), assuming $\mathcal{E}(t)=\mathrm{const}$ and $\gamma_T=\gamma_L=0$. Calculations of the inversion dynamics for different field amplitudes defined by the parameter $\xi=\Omega_R/\Delta\omega$ are shown in Fig. 1. Here, the inversion is plotted as a function of the dimensionless time $T=\Omega_R t/2\pi$. The calculations performed demonstrate that the Rabi frequency strongly depends on the depolarization parameter ξ.

At $\xi\ll 1$ Rabi oscillations are practically absent: $w(t)\sim\mathrm{const}$, that corresponds to weak coupling between QD and electric field: $(\Omega_R\to 0)$. The increase in ξ leads to the appearance of the Rabi oscillations. Following Figs. 2 and 3 demonstrates the inversion calculations for finite values of the parameter δ. Thus, at $\xi=0.2$, small amplitude oscillations of

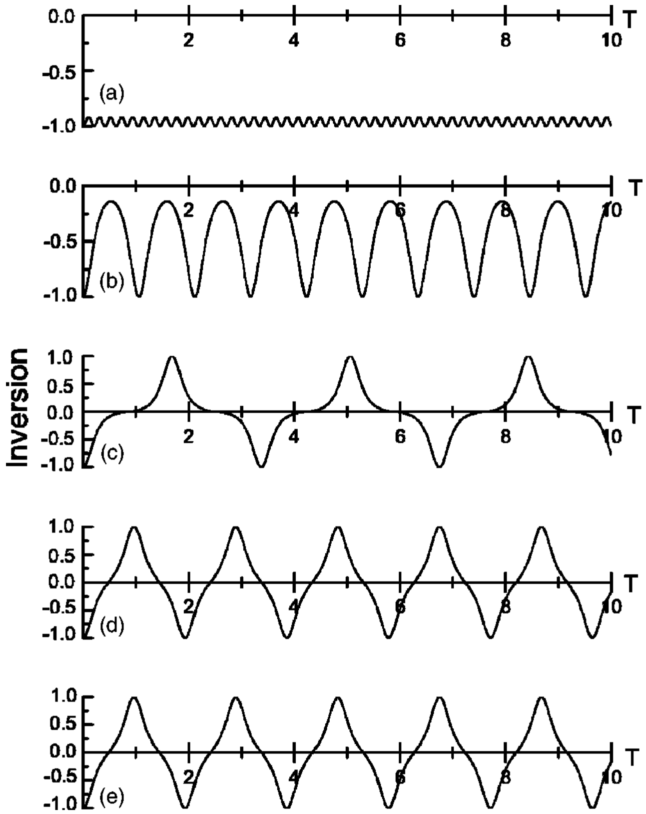

FIG. 2. Rabi oscillations of the inversion, $\delta=-\Delta\omega$: (a) $\xi=0.2$; (b) $\xi=0.495$; (c) $\xi=0.5001$; (d) $\xi=0.51$; (e) $\xi=0.6$.

the inversion are observed [see Fig. 2(a)]. At $\xi=\xi^{cr}=0.5$ *the bifurcation in the oscillation dynamics* is predicted [compare Figs. 2(b) and 2(c)], that separates two oscillatory regimes with drastically different characteristics. In the vicinity of the bifurcation, Rabi oscillations are *essentially anharmonic* [see Figs. 2(c), 2(d)]. This means that in the frequency domain the Mollow triplet[17] (which characterizes the atomic Rabi oscillations) is transformed to a more complicated spectrum that contains satellites corresponding to higher orders of the Rabi oscillations frequency. The anharmonism in the Rabi oscillations disappears at the further increase in ξ; see Fig. 2(e). When $\xi>1$, inversion behavior satisfies the following

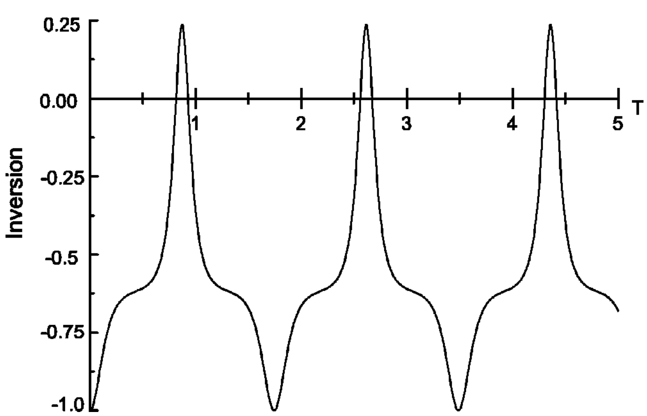

FIG. 3. Rabi oscillations of the inversion. Influence of the detuning ($\delta=-0.5\Delta\omega$; $\xi=0.1502$).

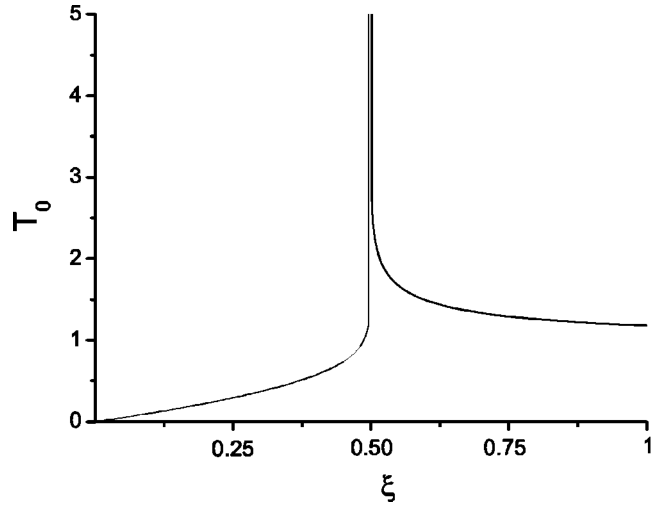

FIG. 4. The period of the Rabi oscillations as a function of the depolarization parameter ξ for $\delta=-\Delta\omega$.

approximation: $w(t)\cong\cos(2\pi T)$, that corresponds to the conventional picture of the Rabi oscillations.[16,17]

In order to explain the bifurcation of the inversion, which results from the influence of nonlinear terms in Eqs. (3) and (4), we analyze the phase portrait of Eqs. (3)–(5). For $\gamma_T=\gamma_L=0$ the system is conservative, and the integral of motion is given by $u^2+v^2+w^2=1$. As a result, from Eqs. (3)–(5) it easily to obtain the expression as follows:

$$v(w)=\frac{1}{2\xi}(w+1)\left(w+2\xi\frac{\delta}{\Omega_R}+1\right), \qquad (7)$$

which describes the phase portrait picture.

The dimensionless period of Rabi oscillations is given by the integral

$$T_0=\frac{1}{\pi}\int_{-1}^{w_0}\frac{dw}{\sqrt{1-w^2-v^2(w)}}, \qquad (8)$$

where $v(w)$ is defined by Eq. (7) and w_0 is the root of algebraic equation $v^2(w)+w^2=1$ closest to $w=-1$. Figure 4 depicts the dependence of the Rabi period T_0 on the depolarization parameter ξ for the case $\delta=-\Delta\omega$. When $\xi=\xi^{cr}=0.5$, $T_0\to\infty$. At small values of ξ there exist small-period Rabi oscillations, while $T_0\approx 1$ at large values of ξ ($\xi>2$) (that is in agreement with previous results presented in Fig. 2). Formulas (7) also shows that the value $\xi^{cr}=0.5$ separates two markedly different oscillatory regimes. When $\xi<0.5$, the inversion of the system cannot cross the axis $w=0$ (because it would contradict the integral of motion), while at $\xi>0.5$ the inversion oscillates in the region $-1<w<1$. Two different synchronism conditions correspond to these two regimes: when $\xi>0.5$ the exact synchronism regime is averaged over the Rabi period; while at $\xi<0.5$ the averaged over the Rabi period detuning takes a finite value that increases with the ξ decrease.

Now, let us investigate the role of the detuning in manifestation of Rabi oscillations. Calculations performed demonstrate *the decrease in* ξ^{cr} with the decrease in $|\delta|$. When

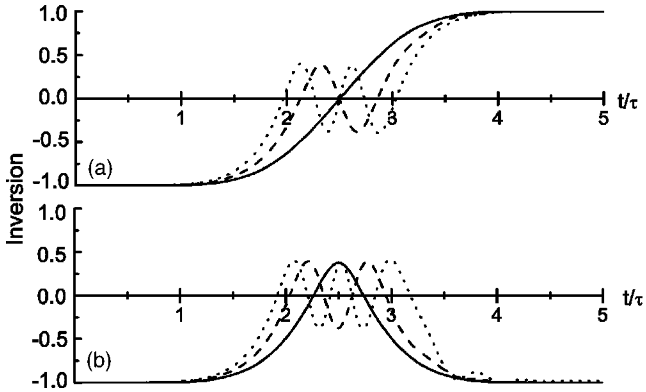

FIG. 5. Rabi oscillation dynamics of QD interacting with the Gaussian pulse: $t_0=0.5\tau$, $\tau\Delta\omega=15$. Two different regimes of the inversion: (a) $\xi=1.0$ (solid line), $\xi=1.26$ (dashed line), $\xi=1.51$ (dotted line); (b) $\xi=1.13$ (solid line), $\xi=1.39$ (dashed line), $\xi=1.61$ (dotted line).

$\delta=-0.9\Delta\omega$, the bifurcation in Rabi oscillations arises at $\xi^{cr}=0.4$, while $\delta=-0.6\Delta\omega$ gives $\xi^{cr}=0.2$. Figure 3 shows the inversion dynamics at $\delta=-0.5\Delta\omega$; in this case $\xi^{cr}=0.15$ and oscillations are essentially anharmonic. Thus, the bifuracation arises in lower fields. It should be noted that the bifurcation disappears at the further decrease $|\delta|$: $\xi^{cr}\to 0$.

Next, we consider a QD interacting with an optical pulse, whose time duration τ is much less than the relaxation times in the system. Numerical calculations for the Gaussian pulse $\mathcal{E}=\mathcal{E}_0\exp[-(t-t_0)^2/\tau^2]$ have been carried out. We consider the Rabi frequency to be associated with the peak field strength of the Gaussian pulse: $\mathcal{E}_0=\mu\Omega_{R0}/\hbar$, so that $\xi=\Omega_{R0}/\Delta\omega$. The inversion dynamics calculation is shown in Fig. 5 for $\delta=-\Delta\omega$. Here, the inversion is plotted as a function of the dimensionless parameter t/τ for different values of ξ. When $\xi\geqslant 1$, inversion demonstrates two different regimes. In the first regime [Fig. 5(a)], the inversion final state w_f is the excited state $(t\to\infty,\ w\to 1)$. In the second regime [Fig. 5(b)], inversion returns back to the ground state $(t\to\infty,\ w\to -1)$. When $\xi\leqslant 1$ (that corresponds to small Rabi frequencies) only the second regime is manifested. It should be emphasized that the spontaneous radiation which is not considered in our model, leads to the decay of the final state of inversion. However, the QD can stay in the excited state for a rather long time. Indeed, the spontaneous radiation life-time of spherical QD[29] can be associated, according to Eq. (80) from Ref. 25, with the resonant frequency shift $\Delta\omega$ by

$$T_{sp}\simeq\frac{1}{4\tau\Delta\omega}\left(\frac{\lambda}{4\pi R\sqrt{\varepsilon_h}}\right)^3. \tag{9}$$

For a GaAs QD with the radius $R\simeq 3$ nm, dielectric constant $\varepsilon_h=12$ and $\tau\Delta\omega=15$ at the wavelength $\lambda=1.3$ μm Eq. (9) gives $T_{sp}\sim 1.3\times 10^2$. This result justifies neglecting in our model of the decay of the inversion final state. Note that the Bohr radius for such QDs is about 10 nm,[28] so that the strong confinement approximation used in our paper is valid. For the QD considered, the estimate $\hbar\Delta\omega\simeq 1$ meV follows from Ref. 25. This corresponds to the value $\hbar\Omega_R\simeq 0.5$ meV for the case $\xi^{cr}=0.5$. Such value of Ω_R is reachable in the pump-probe and microcavities experiments.[30]

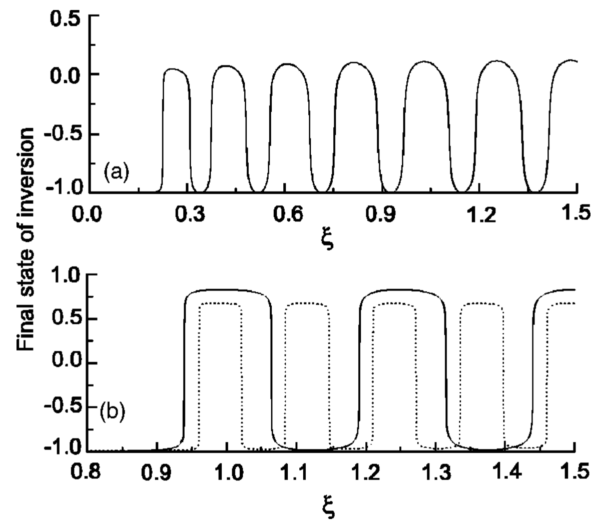

FIG. 6. The final state of inversion as a function of the peak field strength of the Gaussian pulse defined by parameter ξ: (a) undamped system, $\delta=-0.5\Delta\omega$, $\tau\Delta\omega=15$; (b) damped system ($\gamma_T=\gamma_L=10^{-3}\Delta\omega$), $\delta=\Delta\omega$, $\tau\Delta\omega=15$ (solid line) and $\tau\Delta\omega=30$ (dotted line).

Now, let us consider the dependence of the inversion final state on the parameter ξ. Numerical calculations performed when $\delta=-\Delta\omega$ show that at $\xi\geqslant 1$ w_f demonstrates step-like transitions from the ground to the excited state with $w_f=1$. The width of the steps is strongly dependent on the pulse duration. A similar effect for isotropic bulk media has been predicted in Ref. 27. However, the medium in Ref. 27 is considered as an undamped system when carrier field frequency is resonant with atomic transition frequency. The influence of the detuning and damping significantly changes the effect manifestation; see Fig. 6. When $\delta=-0.5\Delta\omega$, step-like transitions start at $\xi>0.25$ and are observed in the region $-1<w_f<0.1$ [Fig. 6(a)]. This result means that the threshold value of the peak pulse strength of the first step-like transition of the inversion can be sufficiently low (at least for four times less as compared with the case $\xi\geqslant 1$). For the case of damped QD, the final state of inversion does not reach a fully inverted state $(w_f=1)$ and does not return back to the ground state $(w_f=-1)$ [Fig. 6(b)]. The latter result qualitatively corresponds to that obtained in Ref. 15, where the damping mechanism has been considered on the basis of the microscopic theory of the QD phonon-electron coupling. However, Ref. 15 does not take local fields into account. Therefore, in order to elaborate a more complete theory describing shapes of that steps, the electron-phonon coupling mechanism developed in Ref. 15 must be incorporated in our local field theory.

Local field theory in anisotropic QDs[23] predicts the polarization dependence of the resonant shift $\Delta\omega$. Therefore, for an anisotropic QD, effects predicted in the current paper are strongly dependent on the incident field polarization that may serve as a methodological basis for their experimental observation.

In conclusion, local fields in QDs are predicted to entail the following effects: (i) The dependence of the Rabi oscillations period on the frequency shift induced by local fields. (ii) The bifurcation in the QD inversion; in the vicinity of that bifurcation Rabi oscillations are essentially anharmonic. (iii) The step-like transitions of the inversion as a function of the ultrashort optical pulse peak strength. The latter result can be used in quantum information processing: as the system has two stable states: "0" and "1," it can be switched from one state to another. Therefore, the array of QDs controlled by an applied field can be used as a basis for logic operations.

The work was partially supported by the NATO Science for Peace Program under Project No. Sfp-972614 and the NATO Linkage Grant under Project No. PST.CLG.980375.

[1] D. Bimberg, M. Grundmann, and N. N. Ledentsov, *Quantum Dot Heterostructures* (Wiley, Chichester, 1999).
[2] N. N. Ledentsov, IEEE J. Sel. Top. Quantum Electron. **8**, 1015 (2002).
[3] A. Ekert and R. Jozsa, Rev. Mod. Phys. **68**, 733 (1996).
[4] P. Chen, C. Piermarocchi, and L. J. Sham, Phys. Rev. Lett. **87**, 067401 (2001).
[5] J. R. Guest, T. H. Stievater, X. Li, Jun Cheng, D. G. Steel, D. Gammon, D. S. Katzer, D. Park, C. Ell, A. Thranhardt, G. Khitrova, and H. M. Gibbs, Phys. Rev. B **65**, 241310(R) (2002).
[6] M. Pelton and Y. Yamamoto, Phys. Rev. A **59**, 2418 (1999);O. Benson and Y. Yamamoto, *ibid.* **59**, 4756 (1999).
[7] M. V. Artemyev, U. Woggon, R. Wannemacher, H. Jaschinski, and W. Langbein, Nano Lett. **1**, 309 (2001).
[8] J. M. Gerard and B. Gayral, Physica E (Amsterdam) **9**, 131 (2001).
[9] P. Michler, A. Imamoglu, M. D. Mason, P. J. Carson, G. F. Strouse, and S. K. Buratto, Nature (London) **406**, 968 (2000); P. Michler, A. Kiraz, C. Becher, W. V. Schoenfeld, P. M. Petroff, Z. Zhang, E. Hu, and A. Imamoglu, Science **290**, 2282 (2000).
[10] C. Santori, M. Pelton, G. Solomon, Y. Dale, and Y. Yamamoto, Phys. Rev. Lett. **86**, 1502 (2001).
[11] A. Thranhardt, C. Ell, G. Khitrova, and H. M. Gibbs, Phys. Rev. B **65**, 035327 (2002).
[12] A. Schulzgen, R. Binder, M. E. Donovan, M. Lindberg, K. Wundke, H. M. Gibbs, G. Khitrova, and N. Peyghambarian, Phys. Rev. Lett. **82**, 2346 (1999).
[13] T. H. Stievater, Xiaoqin Li, D. G. Steel, D. Gammon, D. S. Katzer, D. Park, C. Piermarocchi, and L. J. Sham, Phys. Rev. Lett. **87**, 133603 (2001).
[14] H. Kamada, H. Gotoh, J. Temmyo, T. Takagahara, and H. Ando, Phys. Rev. Lett. **87**, 246401 (2001).
[15] J. Forstner, C. Weber, J. Danckwerts, and A. Knorr, Phys. Rev. Lett. **91**, 127401 (2003).
[16] W. P. Schleich, *Quantum Optics in Phase Space* (Wiley, Berlin, 2001).
[17] M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge, University Press, Cambridge, 1997).
[18] A. A. Budini, R. L. de Matos Filho, and N. Zagury, Phys. Rev. A **67**, 033815 (2003).
[19] M. Kasevish, Science **298**, 1363 (2002).
[20] S. Schmitt-Rink, D. A. B. Miller, and D. S. Chemla, Phys. Rev. B **35**, 8113 (1987).
[21] B. Hanewinkel, A. Knorr, P. Thomas, and S. W. Koch, Phys. Rev. B **55**, 13 715 (1997).
[22] S. A. Maksimenko, G. Ya. Slepyan, V. P. Kalosha, N. N. Ledentsov, A. Hoffmann, and D. Bimberg, Mater. Sci. Eng., B **82**, 215 (2001).
[23] G. Ya. Slepyan, S. A. Maksimenko, V. P. Kalosha, J. Herrmann, N. N. Ledentsov, I. L. Krestnikov, Zh. I. Alferov, and D. Bimberg, Phys. Rev. B **59**, 12 275 (1999).
[24] S. A. Maksimenko, G. Ya. Slepyan, V. P. Kalosha, S. V. Maly, N. N. Ledentsov, J. Herrmann, A. Hoffmann, D. Bimberg, and Zh. I. Alferov, J. Electron. Mater. **29**, 494 (2000).
[25] G. Ya. Slepyan, S. A. Maksimenko, A. Hoffmann, and D. Bimberg, Phys. Rev. A **66**, 063804 (2002).
[26] M. E. Crenshaw and C. M. Bowden, Phys. Rev. Lett. **85**, 1851 (2000);M. E. Crenshaw and C. M. Bowden, Phys. Rev. A **63**, 013801 (2001).
[27] M. E. Crenshaw, M. Scalora, and C. M. Bowden, Phys. Rev. Lett. **68**, 911 (1992).
[28] Y. Fu, M. Willander, and E. L. Ivchenko, Superlattices Microstruct. **27**, 255 (2000).
[29] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, Oxford, 1960).
[30] G. Khitrova, H. M. Gibbs, F. Jahnke, M. Kira, and S. W. Koch, Rev. Mod. Phys. **71**, 1591 (1999).