Matter Coupling to Strong Electromagnetic Fields in Two-Level Quantum Systems with Broken Inversion Symmetry

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(Received 21 July 2008; published 12 January 2009)

We demonstrate theoretically the parametric oscillator behavior of a two-level quantum system with broken inversion symmetry exposed to a strong electromagnetic field. A multitude of resonance frequencies and additional harmonics in the scattered light spectrum as well as an altered Rabi frequency are predicted to be inherent to such systems. In particular, dipole radiation at the Rabi frequency appears to be possible. Since the Rabi frequency is controlled by the strength of the coupling electromagnetic field, the effect can serve for the frequency-tuned parametric amplification and generation of electromagnetic waves. Manifestation of the effect is discussed for III-nitride quantum dots with strong built-in electric field breaking the inversion symmetry. Terahertz emission from arrays of such quantum dots is shown to be experimentally observable.

DOI: 10.1103/PhysRevLett.102.023601 PACS numbers: 42.50.Hz, 47.20.Ky, 78.67.Hc, 85.60.Jb

The resonant interaction of quantum systems with a strong electromagnetic field [1,2] is permanently in the focus of interest, both due to the high methodological value of the arising problems and their direct relation to current applied projects, such as new generation of high-efficient lasers [3], laser cooling of atoms [2], development of basis for quantum information processing [4], etc. One of the bright manifestations of the strong field-matter coupling is the Rabi effect [2]: oscillations of the level population in a quantum system exposed to a monochromatic electromagnetic wave. The simplest physical model leading to harmonic Rabi oscillations is a two-level symmetrical quantum system placed in a given classical single-mode electromagnetic field [2]. The incorporation of additional physical factors into the simplest model results in many nontrivial effects. For example, accounting for the quantum nature of light leads to the concept of radiation-dressed atoms [1] and the "collapse-revivals" phenomenon in the population dynamics of a system exposed to coherent light. Time-domain modulation of the field-matter coupling constant [5], local-field effects in nanostructures [6,7], and phonon-induced dephasing [8] provide new possibilities for the control of the Rabi oscillations. Many interesting effects manifest themselves in more complex systems, such as two coupled Rabi oscillators [9], Rabi oscillators based on superconducting electrical circuits [4], and systems where Rabi oscillations are strongly influenced by intraband motion of quasiparticles [10]. In the given Letter, we investigate the role in the Rabi effect of a new physical factor—violation of the inversion symmetry. Inherent in many quantum systems, the factor is ignored by the conventional physical model [2] of Rabi oscillations. Meanwhile, our theoretical analysis predicts pronounced manifestation of the violation in a set of observable effects in different physical systems.

Let us consider a two-level quantum system with $|a\rangle$ and $|b\rangle$ as excited and ground states, respectively. Let the system interacts with a classical linearly polarized monochromatic electromagnetic field. The system is described by the Hamiltonian $\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_{\text{int}}$. The free-particle Hamiltonian, written in the basis of these two states, is $\hat{\mathcal{H}}_0 = \hbar\omega_0\hat{\sigma}_z/2$, where $\hat{\sigma}_z$ is the Pauli matrix and ω_0 is the resonant frequency of the two-level system. The interaction Hamiltonian is expressed in terms of the amplitude $\mathbf{E}$ and frequency ω of the driving field by $\hat{\mathcal{H}}_{\text{int}} = -\mathbf{E}\hat{\mathbf{d}}\cos(\omega t)$, where $\hat{\mathbf{d}}$ is the electric dipole moment operator and $\mathbf{d}_{ij} = \langle i|\hat{\mathbf{d}}|j\rangle$ are its matrix elements. The critical assumption, which distinguishes the systems being considered from standard ones, is the violation of the inversion symmetry. Since in that case the states $|a\rangle$ and $|b\rangle$ do not possess a certain spatial parity, the diagonal matrix elements of the dipole moment operator prove to be nonequivalent, $\mathbf{d}_{aa} \neq \mathbf{d}_{bb}$, dictating thus physical effects described hereafter.

We shall seek the solution of the Schrödinger equation with the Hamiltonian described above in the form of $|\psi\rangle = C_a(t)|a\rangle + C_b(t)|b\rangle$. Substituting this expression into the Schrödinger equation, we arrive at the equations

$$i\hbar\dot{C}_a = \left[\frac{\hbar\omega_0}{2} - \mathbf{E}\mathbf{d}_{aa}\cos(\omega t)\right]C_a - \mathbf{E}\mathbf{d}_{ab}\cos(\omega t)C_b, \tag{1}$$

$$i\hbar\dot{C}_b = \left[-\frac{\hbar\omega_0}{2} - \mathbf{E}\mathbf{d}_{bb}\cos(\omega t)\right]C_b - \mathbf{E}\mathbf{d}_{ba}\cos(\omega t)C_a. \tag{2}$$

In order to solve these equations with respect to C_a and C_b, we first rewrite them for modified amplitudes $c_{a,b} = C_{a,b}\exp[\pm i\omega_0 t/2 - i\phi_{a,b}(\omega, t)]$, where $\phi_j(\omega, t) = \mathbf{E}\mathbf{d}_{jj}\sin(\omega t)/\hbar\omega$, and signs $\pm$ correspond to indexes a and b, respectively. The system of Eqs. (1) and (2) is then reduced to the form as follows

$$\hbar\,\dot{c}_a = i\mathbf{E}^*_{\rm eff}\mathbf{d}_{ab}\cos(\omega t)c_b e^{i\omega_0 t}, \tag{3}$$

$$\hbar\,\dot{c}_b = i\mathbf{E}_{\rm eff}\mathbf{d}_{ba}\cos(\omega t)c_a e^{-i\omega_0 t}, \tag{4}$$

where the effective electric field strength is

$$\mathbf{E}_{\rm eff}(t) = \mathbf{E}e^{-i\kappa\sin(\omega t)} = \mathbf{E}\sum_{n=-\infty}^{\infty} J_n(\kappa)e^{in\omega t}, \tag{5}$$

$J_n(\kappa)$ is the Bessel function of the first kind, and $\kappa = \mathbf{E}(\mathbf{d}_{bb} - \mathbf{d}_{aa})/\hbar\omega$ is the symmetry violation parameter. Let us stress that Eqs. (3) and (4) are analogous to standard equations of two-level system [2] with the only difference that the driving field amplitude $\mathbf{E}$ is replaced by the effective amplitude (5). That the effective amplitude $\mathbf{E}_{\rm eff}(t)$ is time-dependent causes the system to become a parametric oscillator with full set of intrinsic properties, unusual for standard Rabi oscillators. In particular, a multitude of resonant frequencies $\omega = \omega_0/n, n = 1, 2, 3, \ldots$ appears in the system. Further, we shall assume that the driving field frequency ω is in the vicinity of the frequency ω_0/m (m-th resonance) and the conditions $|\mathbf{E}\mathbf{d}_{ab}mJ_m(\kappa)/\kappa\hbar(\omega_0 - m\omega)| \gtrsim 1$ and $|\mathbf{E}\mathbf{d}_{ab}nJ_n(\kappa)/\kappa\hbar(\omega_0 - n\omega)| \ll 1 (n \neq m)$ are fulfilled, allowing us to neglect interaction of the two-level system with all harmonics (5) other than harmonics $m \pm 1$. Then Eqs. (3) and (4) resume the form analogous to equations for symmetric two-level systems and can easily be solved by invoking the rotating-wave approximation [2]. Assuming the nondiagonal dipole matrix elements to be real valued, we obtain

$$C_a(t) = \left\{C_a(0)\left[\cos\left(\frac{\Omega t}{2}\right) - \frac{i\Delta}{\Omega}\sin\left(\frac{\Omega t}{2}\right)\right] + i\frac{\Omega_R}{\Omega}C_b(0)\sin\left(\frac{\Omega t}{2}\right)\right\}e^{-im\omega t/2}e^{i\phi_a(\omega,t)}, \tag{6}$$

$$C_b(t) = \left\{C_b(0)\left[\cos\left(\frac{\Omega t}{2}\right) + \frac{i\Delta}{\Omega}\sin\left(\frac{\Omega t}{2}\right)\right] + i\frac{\Omega_R}{\Omega}C_a(0)\sin\left(\frac{\Omega t}{2}\right)\right\}e^{im\omega t/2}e^{i\phi_b(\omega,t)}, \tag{7}$$

where the parameters $\Delta = \omega_0 - m\omega$, $\Omega = \sqrt{\Omega_R^2 + \Delta^2}$, and the Rabi frequency

$$\Omega_R = 2\mathbf{E}\mathbf{d}_{ab}mJ_m(\kappa)/\kappa\hbar \tag{8}$$

are written in the vicinity of m-th resonance. Since we consider the strong coupling regime, the driving field $\mathbf{E}$ is assumed to be sufficiently strong, $\Omega_R\tau \gg 1$, to neglect impact of the linewidth $\hbar/\tau$. Note that the Rabi frequency in systems with broken inversion symmetry (8) shows nonmonotonic dependence on the driving field strength and even can turn zero at those values of $\mathbf{E}$ which correspond to roots of the Bessel function $J_m(\kappa)$.

It is well known that in symmetrical two-level systems, the Rabi effect manifests itself in the power spectrum of the scattered light by peaks centered at the incident light frequency ω and at the displaced frequencies $\omega \pm \Omega$ (Mollow triplet, [11]). In order to reveal peculiarities of the scattered light induced by the symmetry violation, it is suffice to consider the process in the framework of classical electrodynamics. It allows analyzing electronic subsystem, in respect to irradiation of electromagnetic waves, as a classical dipole with the oscillating dipole moment $\mathbf{d}(t) = \langle\psi|\hat{\mathbf{d}}|\psi\rangle$. For definiteness, we identify initial time as that corresponding to system being in the excited state, i.e., $C_a(0) = 1$ and $C_b(0) = 0$. Then, substituting wave function $|\psi\rangle$ with coefficients (6) and (7) into expression for the dipole moment and omitting the time-independent terms, we arrive at the expression

$$\mathbf{d}(t) = (\mathbf{d}_{aa} - \mathbf{d}_{bb})\frac{\Omega_R^2}{4\Omega^2}e^{i\Omega t} - \mathbf{d}_{ab}\frac{\Omega_R}{2\Omega}\sum_{n=-\infty}^{\infty} J_{m-n}(\kappa)e^{in\omega t} \times\left[\frac{\Delta}{\Omega} + \frac{1}{2}\left(1 - \frac{\Delta}{\Omega}\right)e^{-i\Omega t} - \frac{1}{2}\left(1 + \frac{\Delta}{\Omega}\right)e^{i\Omega t}\right] + \text{c.c.} \tag{9}$$

for the dipole moment in the vicinity of m-th resonance. As follows from Eq. (9), the radiation spectrum consists of a singlet at the frequency Ω, and an infinite sequence of triplets with the frequencies $n\omega$, $n\omega \pm \Omega$ (see Fig. 1). It should be emphasized that the frequency multiplication is the general property of systems with broken inversion symmetry and can be used for the high harmonic generation [12]. Amplitudes of harmonics of the triplets rapidly decrease with increasing n or m, while the singlet Ω amplitude depends on neither n nor m. Radiation of the dipole (9) in the vicinity of each resonance is characterized by its own Rabi frequency (8) decreasing with the resonance number m increase. In systems with inversion symmetry, diagonal elements of the dipole moment are identical. As a result, higher resonances ($m > 1$), higher triplets ($n > 1$), and the singlet vanish. In that case, coefficients (6) and (7) and expression for the dipole moment (9) coincide with the solution of the problem presented in Ref. [2].

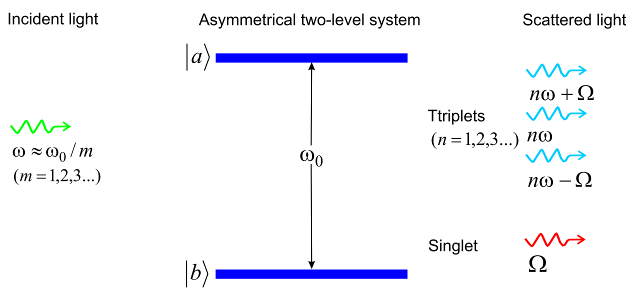

FIG. 1 (color online). Schematics of the light scattering in a two-level system with broken inversion symmetry in the vicinity of one of the possible resonances $\omega = \omega_0/m$.

Further, we confine consideration to systems with a weak violation of the inversion symmetry, when the condition $|\kappa| \ll 1$ holds true. In such systems, the asymmetry effect is expected to be most pronounced in the vicinity of the first resonance $m = 1$ ($\omega \approx \omega_0$). If then we restrict ourselves to the most interesting case of resonant driving field, when $\Delta = 0$ and $\Omega = \Omega_R$, we reduce (9) to the expression

$$\mathbf{d}(t) = \frac{\mathbf{d}_{aa} - \mathbf{d}_{bb}}{2} \cos(\Omega_R t) - \mathbf{d}_{ab} \sin(\omega_0 t) \sin(\Omega_R t), \tag{10}$$

where Rabi frequency (8) takes its conventional form $\Omega_R = \mathbf{E}\mathbf{d}_{ab}/\hbar$. In typical quantum systems, the Rabi frequency is much less then the driving field frequency $\omega = \omega_0$. Therefore, in addition to the high-frequency harmonics $\omega_0 \pm \Omega_R$ [second term in the right-hand part of Eq. (10)], violation of the inversion symmetry leads to the irradiation of low-frequency electromagnetic waves at Rabi frequency [first term in the right-hand part of Eq. (10)] with the time-averaged radiation intensity

$$I_R = \frac{|\mathbf{d}_{aa} - \mathbf{d}_{bb}|^2}{12c^3} \Omega_R^4. \tag{11}$$

The quantity $|\mathbf{d}_{aa} - \mathbf{d}_{bb}|$ is further referred to as effective dipole moment. Note that the frequency of the scattered radiation Ω_R depends only on the driving field $\mathbf{E}$ and does not depend on both the frequency of the scattering system ω_0 and the frequency of incident light ω.

The simplest quantum system devoid of the inversion center is a Rydberg hydrogen atom imposed to a homogeneous static electric field $\mathcal{E}$. Assuming the atom to be in ground state, we find that the Rabi frequency is expressed in terms of the Bohr radius $a_B = \hbar^2/m_e e^2$ by $\Omega_R = 4(2/3)^5 e a_B E/\hbar$. Correspondingly, for the electric field $\mathcal{E}$ much less than the intra-atomic electric field e/a_B^2, the intensity of the radiation at the Rabi frequency is determined by Eq. (11) with the effective dipole moment given by $|\mathbf{d}_{aa} - \mathbf{d}_{bb}| = (1/8)(4/3)^{11} a_B^3 \mathcal{E}$. This expression is applicable not only to hydrogen atom but also can be used for estimating parameters of the emission at the Rabi frequency in arbitrary quantum systems with broken inversion symmetry. For that aim, the Bohr radius a_B should be replaced by the characteristic linear extension of the system. Therefore, the radiation intensity (11) rises with the increase of the system size. In that connection, confined semiconductor nanostructures with discrete energy spectrum and linear extension multiply exceeding the atomic size, quantum dots (QDs) [3], can serve as prospective systems for the effect observation.

In quantum dots, driving electromagnetic field transfers electrons from valence band into conduction band; this is why in QDs the ground state $|b\rangle$ corresponds to the absence of free carriers while first excited state $|a\rangle$ is the state with electron in the conduction band and hole in the valence band. In that case, $\hbar\omega_0$ is approximately equal to the semiconductor bandgap, and the Rabi frequency is determined by the standard expression $\Omega_R = \mathbf{E}\mathbf{d}_{cv}/\hbar$ with the dipole matrix element $\mathbf{d}_{cv}$ corresponding to interband transitions. Among a variety of different types of QDs, nitride-based confined structures seem to be most promising for the effect observation (for parameters of III-nitrides AlN, GaN, and InN see Ref. [13]). Indeed, GaN and similar III-group nitride alloys have a hexagonal (wurtzite) structure. As a consequence of giant piezoelectric effect inherent in hexagonal crystals, QDs based on structures AlN/GaN and InN/GaN have a strong built-in strain-induced electric field with strength $\mathcal{E}$ of several MV/cm [14,15]. Because of the strong electric field, conduction-band electrons and valence-band holes in the QDs get spatially separated [16,17], and the effective dipole moment of a III-nitride QD is given by simple relation $|\mathbf{d}_{aa} - \mathbf{d}_{bb}| \sim el$, where l is the QD height. Then, for typical l of several nanometers, the effective dipole moment of nitride QDs is estimated as $|\mathbf{d}_{aa} - \mathbf{d}_{bb}| \sim 10$ Debye, what is tens thousands times as large as the effective dipole moment of hydrogen atom in the same electric field.

Since the Rabi frequency depends on the driving field strength $\mathbf{E}$, the broken inversion symmetry-induced low-frequency singlet Ω_R in the dipole emission spectrum can be used for the tunable generation of electromagnetic waves at Rabi frequency with intensity (11). This problem is especially challenging for frequency ranges where traditional methods either fail or inefficient, such as terahertz domain. Since this domain lies between radio and optical frequency ranges, neither optical nor microwave techniques are directly applicable for generating THz waves. Therefore, a search for effective THz radiation sources is one of most excited fields of modern applied physics [18–20]. One of the latest trends to fill the THz gap is using nanostructures as THz emitters and detectors. The quantum cascade THz transitions in QD systems [21,22] and different electron mechanisms of THz emission from carbon nanotubes [23–25] have been proposed and are being actively studied. Thus, the proposed mechanism of THz emission fits well the current tendencies in nanophotonics.

Parameters of the THz emission from III-nitride QDs can be evaluated in the following way. Using the estimate $d_{cv} \sim 10$ Debye for the QD interband dipole moment [13], we obtain that the Rabi frequency turns out to be lying in the THz range at relatively weak strength of the driving field, $E \sim 10^5$ V/cm. Then, substituting the previously obtained value of the effective dipole moment $|\mathbf{d}_{aa} - \mathbf{d}_{bb}| \sim 10$ Debye into Eq. (11), for the typical QD area $\sim 10^{-12}$ cm^2, we arrive at the radiative intensity per single QD $\sim 10^{-11}$ W/cm^2 in the THz range. This estimate multiples as N^2 for array of N identical QDs with the lateral extension less than the Rabi wavelength $\lambda_R = 2\pi c/\Omega_R$. In such an array, all QDs emit waves in phase. The THz-range wavelength $\lambda_R \sim 10^{-2}$ cm restricts the array lateral size. Taking into account that the typical density of nitride QDs is $\sim 10^{11}$ cm^{-2}, we can estimate the number of QDs in such an array by $N \sim 10^7 - 10^8$. Therefore, the THz emission power from the submillimeter-sized QD array may approach the micro-Watt level, which is characteristic for the resonant-tunneling diodes based on carbon nanotubes [26]. Certainly, state-of-the-art THz quantum cascade lasers demonstrate essentially larger output power [27]. However, array of QDs with broken inversion symmetry provides smooth frequency tuning by the variation of the driving field intensity. The reducing impact of inhomogeneous broadening is beyond the estimate and will be considered elsewhere. As to the homogeneous mechanisms of the line broadening, like phonon scattering, they are accounted for phenomenologically in the linewidth $\hbar/\tau$ and do not impose any specific restrictions on the observability of the effect, but just influence the criterion of the strong light-matter coupling regime. Thus, the upper estimate presented of the radiation output allows proposing the broken inversion symmetry-induced mechanism of the radiation for the development of novel-type THz emitters based on QDs avoid inversion symmetry. Obviously, parametric amplification of THz radiation in QD arrays is also possible and can be applied for THz detecting.

One more type of asymmetrical two-level quantum systems where the discussed effect can be observed is superconducting quantum qubits [4,28]. Away from the charge degeneracy point of the superconducting quantum circuits [4] formed by Josephson qubits in microstrip resonators, the qubit symmetry is broken, and the Hamiltonian of the system (see Eq. (16) in Ref. [4]) takes the form analogous to that used in our analysis. At that, the resonant transition frequency amounts to ~10 GHz while the Rabi frequency lies in the range ~100 MHz. Intensity of low-frequency line in the spectrum of Rabi oscillations can be controlled by the changing of dc gate voltage. Chiral nanostructures, including chiral carbon nanotubes [29], should also be noted as prospective systems for observation of the effect, since the chirality breaks the inversion symmetry. New interesting effects are expected for asymmetric two-level oscillators placed inside band-gap structures like photon crystal, microcavity, or nanoantenna. For example, by corresponding manipulation of the photonic crystal parameters, the frequencies ω and ω_0 can be chosen laying in the band gap while the frequency Ω lies in the transparency band. In that case, the strong coupling regime is realized for the pump field, while at the frequency Ω the photonic crystal serves as an antenna transforming near field into far field. Similar effect appears in microcavity with resonant frequency close to ω and leaking modes at the frequency Ω.

In the Letter, we have assumed that the incident light pulse is much longer than periods of both incident and scattered electromagnetic field. When a collection of oscillators with broken inversion symmetry is illuminated by an extremely short pulse, the difference between amplitudes E and E_{eff} in Maxwell-Bloch equations may result in the failure of the area theorem and in the related effect of the carrier-wave Rabi flopping. Such an effect is observed in semiconductors when the Rabi frequency becomes comparable to the band-gap frequency [30].

To conclude, let us stress that violation of the inversion symmetry is common property of quantum oscillators of different physical origination and, consequently, the effect predicted is expected to manifest itself in spectral characteristics of different optical processes, e.g., resonant fluorescence of molecules.

The research was partially supported by the INTAS project 05-1000008-7801, IB BMBF (Germany) project BLR 08/001, the EU FP7 TerACaN project FP7-230778, RFBR (Russia) project 08-02-90004, "Development of Scientific Potential of Russian Higher Education" Program (project 2.1.2/2115), and BRFFR (Belarus) project F08R-009. The work of O. V. K. and S. A. M. was partially carried out during the stay at the Institut für Festkörperphysik, TU Berlin, and supported by DAAD and DFG, respectively.

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