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Population inversion in two-level systems possessing permanent dipoles

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Bare-state population inversion is demonstrated in a two-level system with all dipole matrix elements nonzero. A laser field is resonantly driving the sample, whereas a second weaker and lower frequency coherent field additionally pumps it near resonance with the dynamically Stark-splitted states. Due to the existence of differing permanent dipole moments in the excited and ground bare states, quantum coherences among the involved dressed states are induced, leading to inversion in the steady state. Furthermore, large refractive indices are feasible, as well as the determination of the diagonal matrix elements via the absorption or emission spectra. The results apply to available biomolecular, spin, or asymmetric quantum dot systems.

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I. INTRODUCTION

Population inversion in a few-energy-level quantum system is strongly linked with its optical response and has resulted in traditional masing or lasing effects being successfully demonstrated [1–3]. Subsequently, enormous economical and technological progress was triggered due to quantum electronics. However, at present, atomic steady-state population inversion is achievable essentially only in pumped two-level atomic systems involving efforts via extra transitions with additional photon sources or environmental vacuum modifications [4]. Additionally, two-level quantum dot systems may also exhibit population inversion due to extra phonon-induced decay rates [5]. Meanwhile, lasing without population inversion operates as well with the help of induced quantum coherences [6]. Moreover, free-electron laser sources involving inversions of momentum states exist at higher frequencies, leading to original effects [7].

Over the past decade a range of experiments were performed demonstrating quantum effects in biological samples [8]. Particularly, energy transfer through quantum coherence in photosynthetic systems [9] was observed [10,11], while long-living quantum coherences survive in biological complexes even under normal conditions at room temperature [12]. Quantum coherence and entanglement in the processes of magnetoreception of the surrounding magnetic field [13,14] were examined, too [15]. The single-molecule fluorescence spectroscopy and the emitted photon quantum statistics are further excellent tools for research in quantum biology [16,17]. Furthermore, coherent control of an effective two-level system in a non-Markovian biomolecular environment was investigated [18]. Additional quantum effects in biochemical systems are discussed, for instance, in [19]. An important issue raised in this context is to image tiny objects, such as biological cells or organic molecules, and therefore highly refractive biological media are required [20].

Inspired by these remarkable advances in quantum biochemistry, here, we put forward a realistic scheme that enables the creation of population inversion in certain biological samples acting as two-level systems with all possible dipole matrix elements being nonzero. The effect occurs due to induced quantum coherences which arise from the difference of the permanent dipole moments in the excited and ground states, respectively. This may allow for lasing or amplifying, as well as optical switching devices in biomolecular materials. Large index of refractions without absorption are also feasible, which may lead to an enhanced optical imaging resolution of the biomolecular sample due to a reduced probe-field wavelength inside the medium. Both the absorption or emission spectra can be used to extract the values of permanent dipoles.

The article is organized as follows. In Sec. II we describe the analytical approach and the system of interest, whereas in Sec. III we analyze the obtained results. A summary is given in Sec. IV.

II. THEORETICAL FRAMEWORK

We consider a two-level system possessing permanent dipoles and interacting with two external coherent laser fields. The first laser is near resonance with the transition frequency of the two-level sample, while the second one is close to resonance with the dressed-frequency splitting due to the first laser, respectively (see Fig. 1). As a concrete system, we may consider gamma-globulin macromolecules [19,21] with the transition frequency $\omega_{21} \approx 4.8 \times 10^{15}$ Hz, transition dipole moment $d \approx 1$ D, and the difference between the diagonal dipole moments in the upper and lower bare states given by $|d_{22} - d_{11}| \approx 100$ D. However, the analytical formalism applies equally to spin [22], asymmetric semiconductor quantum dot [23], or other alternative [24] systems promising wider applications. The Hamiltonian describing such a model, in a frame rotating at the first laser frequency ω_L and in the dipole approximation is

$$H = \sum_k \hbar(\omega_k - \omega_L)a_k^\dagger a_k + \hbar\Delta S_z + \hbar\Omega(S^+ + S^-) + \hbar G S_z \cos(\omega t) + i\sum_k (\vec{g}_k \cdot \vec{d})(a_k^\dagger S^- - a_k S^+). \qquad (1)$$

In the Hamiltonian (1) the first three components are, respectively, the free energies of the environmental electromagnetic

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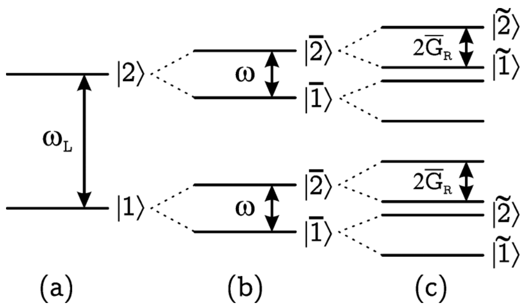

FIG. 1. Energy diagram of a two-level emitter with nonzero values of all involved dipole matrix elements $d_{\alpha\beta}$, $\{\alpha,\beta = 1,2\}$. (a) A moderately intense laser field of frequency ω_L resonantly interacts with the molecular sample, leading to dynamic Stark splitting of its energy levels. (b) A second coherent source of frequency ω, close to the generalized Rabi frequency due to the first laser, is applied, leading to transitions among the dressed states. (c) The "double dressed states," with $2\bar{G}_R$ being the corresponding Rabi splitting frequency.

vacuum modes and molecular subsystems together with the laser-molecule interaction Hamiltonian. There, $\Omega = dE_1/(2\hbar)$ is the corresponding Rabi frequency, with $d \equiv d_{21} = d_{12}$ being the transition dipole moment, while E_1 is the amplitude of the first laser field.

The fourth term accounts for the second laser interacting at frequency ω and amplitude E_2 with the molecular system due to presence of permanent dipoles incorporated in G, i.e., $G = (d_{22} - d_{11})E_2/\hbar$. The last term describes the interaction of the molecular subsystem with the environmental vacuum modes of the electromagnetic field reservoir. Further, $\vec{g} = \sqrt{2\pi\hbar\omega_k/V}\vec{e}_\lambda$ is the molecule-vacuum coupling strength, with $\vec{e}_\lambda$ being the photon polarization vector and $\lambda \in \{1,2\}$, whereas V is the quantization volume. $\Delta = \omega_{21} - \omega_L$ is the laser field detuning from the molecular transition frequency ω_{21}. The molecule bare-state operators $S^+ = |2\rangle\langle 1|$ and $S^- = [S^+]^\dagger$ obey the commutation relations $[S^+,S^-] = 2S_z$ and $[S_z,S^\pm] = \pm S^\pm$. Here, $S_z = (|2\rangle\langle 2| - |1\rangle\langle 1|)/2$ is the bare-state inversion operator. $|2\rangle$ and $|1\rangle$ are, respectively, the excited and ground state of the molecule, while $a_k^\dagger$ and a_k are the creation and the annihilation operator of the k_{th} electromagnetic field mode and satisfy the standard bosonic commutation relations, namely, $[a_k,a_{k'}^\dagger] = \delta_{kk'}$ and $[a_k,a_{k'}] = [a_k^\dagger,a_{k'}^\dagger] = 0$ [25–28]. Notice that the Hamiltonian (1) is incomplete. The following term,

$$\tilde{H} = \hbar\tilde{\Omega}S^+e^{i\omega_L t}\cos(\omega t) + \hbar\tilde{G}S_z\cos(\omega_L t)/2 + \text{H.c.}, \quad (2)$$

will not be taken into account. Here, the first term describes the interaction of the second laser with the molecular system due to the transition dipole d, while the second one represents the interaction of the first laser with the molecule because of permanent dipoles and, hence, $\tilde{\Omega} = dE_2/\hbar$, whereas $\tilde{G} = (d_{22} - d_{11})E_1/\hbar$. Since we assume realistic conditions that $\tilde{\Omega} \ll \omega_L \pm \omega$ [27,28] as well as $\{\tilde{G},\omega\} \ll \omega_L$, the Hamiltonian (2) can be considered as fast oscillating and therefore, neglectable.

In the following, we shall consider a regime where the generalized Rabi frequency $\bar{\Omega} = \sqrt{\Omega^2 + (\Delta/2)^2}$ is larger than the single-molecule spontaneous decay rate as well as the coupling due to permanent dipoles, i.e., $\bar{\Omega} \gg \gamma$ and $\bar{\Omega} > G$. In this case it is more convenient to describe our system in the semiclassical laser-molecule dressed-state picture due to the first applied laser:

$$|2\rangle = \cos\theta|\bar{2}\rangle - \sin\theta|\bar{1}\rangle \text{ and } |1\rangle = \cos\theta|\bar{1}\rangle + \sin\theta|\bar{2}\rangle, \quad (3)$$

with $\tan 2\theta = 2\Omega/\Delta$. Here $|\bar{2}\rangle$ and $|\bar{1}\rangle$ are the corresponding upper and lower dressed states, respectively [see Fig. 1(b)]. Applying the dressed-state transformation to the Hamiltonian (1), one arrives at the following Hamiltonian represented in a frame rotating also at the second laser field frequency, i.e., ω:

$$\begin{aligned} H = {} & \sum_k \hbar(\omega_k - \omega_L)a_k^\dagger a_k + \hbar\bar{\Delta}R_z - \hbar\bar{G}(R^+ + R^-) \\ & + i\sum_k(\vec{g}_k\cdot\vec{d})\{a_k^\dagger(\sin 2\theta R_z/2 + \cos^2\theta R^- e^{-i\omega t} \\ & - \sin^2\theta R^+ e^{i\omega t}) - \text{H.c.}\}, \end{aligned} \quad (4)$$

where $\bar{\Delta} = \bar{\Omega} - \omega(1 - \bar{G}^2/\omega^2)/2 \equiv \bar{\Omega} - \omega/2$ and $\bar{G} = (G/4)\sin 2\theta$, and we have performed the rotating wave approximation with respect to ω, i.e., we have assumed that $\omega \gg \bar{G}$. Eliminating the vacuum modes of the electromagnetic field reservoir in the usual way, by adopting the Born-Markov approximations [25–28] one arrives then at the following dressed-state master equation:

$$\begin{aligned} & \dot{\rho}(t) + i[\bar{\Delta}R_z - \bar{G}(R^+ + R^-),\rho] \\ & = -\frac{\gamma_0}{4}\sin^2 2\theta[R_z,R_z\rho] - \gamma_+\cos^4\theta[R^+,R^-\rho] \\ & \quad - \gamma_-\sin^4\theta[R^-,R^+\rho] + \text{H.c.} \end{aligned} \quad (5)$$

Here $\gamma_{0,\pm}$ are the single-qubit spontaneous decay rates corresponding to dressed-state frequencies ω_L and $\omega_L \pm 2\bar{\Omega}$, respectively. The new quasi-spin operators, i.e., $R^+ = |\bar{2}\rangle\langle\bar{1}|$, $R^- = [R^+]^\dagger$, and $R_z = |\bar{2}\rangle\langle\bar{2}| - |\bar{1}\rangle\langle\bar{1}|$, are operating in the dressed-state picture. They obey the following commutation relations: $[R^+,R^-] = R_z$ and $[R_z,R^\pm] = \pm 2R^\pm$. Notice that in the above master equation, we have neglected the rapidly oscillating terms in the spontaneous emission part, an approximation valid when $\{\bar{\Omega},\omega\} \gg \{\gamma_0,\gamma_\pm\}$.

In the following section we discuss our results, i.e., the possibility to create bare-state population inversion as well as high refractive media.

III. RESULTS AND DISCUSSION

One can observe from Eq. (5) that permanent dipoles lead to the appearance of a pumping term among the dressed states. This term contributes to a completely unexpected behavior of the two-level system. In particular, as we shall see, it can induce pumping the system into an inverted state. The system of equations for the dressed-state inversion and dressed-state polarization operators can be obtained from the master equation (5), namely,

$$\begin{aligned} \langle\dot{R}_z\rangle &= -2i\bar{G}(\langle R^-\rangle - \langle R^+\rangle) - 2\Gamma_+\langle R_z\rangle + 2\Gamma_-, \\ \langle\dot{R}^+\rangle &= (2i\bar{\Delta} - \Gamma)\langle R^+\rangle + i\bar{G}\langle R_z\rangle, \end{aligned} \quad (6)$$

with $\langle R^-\rangle = [\langle R^+\rangle]^\dagger$. Here, $\Gamma_\pm = \gamma(\sin^4\theta \pm \cos^4\theta)$, $\Gamma = \Gamma_+ + \gamma\sin^2 2\theta$, and we have considered that $\gamma_0 = \gamma_\pm \equiv \gamma$, which is the case for a free-space setup. The mean value of

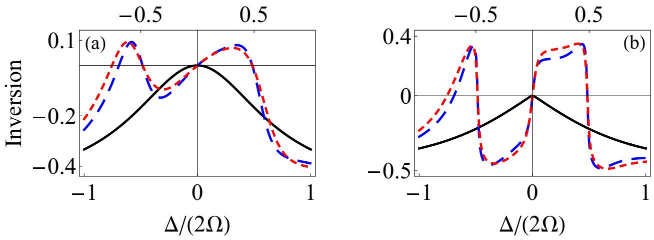

FIG. 2. (Color online) The steady-state dependence of the bare-state inversion operator $\langle S_z\rangle/N$ versus the scaled parameter $\Delta/(2\Omega)$. The solid line is plotted for $G/\gamma = 0$, the long-dashed one is for $G/\gamma = 16$, while the short-dashed curve corresponds to $G/\gamma = 24$. Other parameters are $\Omega/(N\gamma) = 45$ and $\omega/(N\gamma) = 100$, with $\gamma \approx 2.6$ MHz as feasible for gamma globulin. (a) $N = 1$ whereas (b) $N = 50$, while molecules are dense enough to allow for collectivity.

the bare-state inversion operator $\langle S_z\rangle$ can be represented via dressed-state operators as follows:

$$\langle S_z\rangle = \cos 2\theta\langle R_z\rangle/2 - \sin 2\theta(\langle R^+\rangle + \langle R^-\rangle)/2. \quad (7)$$

From the system of Eqs. (6) one immediately obtains the following steady-state relations:

$$\langle R^+\rangle = i\bar G\langle R_z\rangle/(\Gamma - 2i\bar\Delta) \text{ and } \langle R^-\rangle = [\langle R^+\rangle]^\dagger. \quad (8)$$

By inserting Eq. (8) in Eq. (7) one arrives at

$$\langle S_z\rangle = (\cos 2\theta + 4\bar\Delta\bar G\sin 2\theta/[\Gamma^2 + (2\bar\Delta)^2])\langle R_z\rangle/2, \quad (9)$$

where, again, from Eqs. (6) one has that

$$\langle R_z\rangle = 2\Gamma_-/[2\Gamma_+ + (2\bar G)^2\Gamma/(\Gamma^2 + (2\bar\Delta)^2)]. \quad (10)$$

An interesting result here is the nonzero value for the dressed-state coherences $\langle R^\pm\rangle$ [see Eq. (8)]. In the absence of permanent dipoles, i.e., $G = 0$, these quantities are zero in the moderately intense pumping regime considered here. Therefore new physics is expected due to existence of permanent dipoles in bichromatic pumping fields. In particular, Fig. 2(a) shows the mean value of the single-molecule bare-state inversion operator for particular parameters of interest. Steady-state inversion in the bare states is achieved when $\langle S_z\rangle > 0$, and it occurs in the presence of permanent dipoles. On the other hand, Fig. 3(a) depicts the real part of the mean value of the dressed-state coherence operator $\langle R^+\rangle$ in steady state. The minima observed in these behaviors correspond to an inverted molecular bare-state system [compare Figs. 2(a) and 3(a)]. Thus inversion occurs due to the real part of the dressed-state coherences, which can be nonzero in our system.

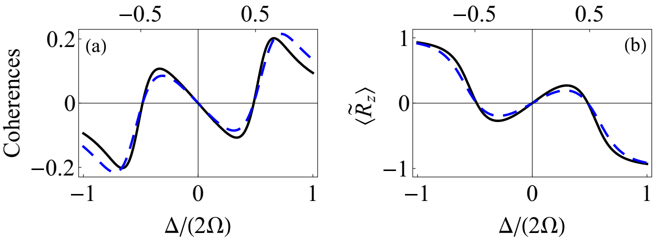

FIG. 3. (Color online) The single-molecule steady-state behaviors of (a) the real part of the dressed-state coherences $\langle R^+\rangle$ and (b) the double dressed-state inversion operator $\langle\tilde R_z\rangle$ versus the scaled detuning $\Delta/(2\Omega)$. The solid line is plotted for $G/\gamma = 16$, while the dashed curve corresponds to $G/\gamma = 24$. Here $\Omega/\gamma = 45$ and $\omega/\gamma = 100$.

One can apply the double dressed-state formalism (see Fig. 1) in order to obtain further information on our system, namely,

$$|\bar 2\rangle = \cos\phi|\tilde 2\rangle + \sin\phi|\tilde 1\rangle, \quad |\bar 1\rangle = \cos\phi|\tilde 1\rangle - \sin\phi|\tilde 2\rangle. \quad (11)$$

This approach is particularly useful to account for vacuum-induced collective effects among the emitters and their corresponding influences on molecular dynamics. By introducing Eq. (11) in the Hamiltonian (4) and, again, eliminating the degrees of freedom related with the environmental vacuum modes in the Born-Markov approximations, one arrives at the double dressed master equation:

$$\dot\rho(t) + i\bar G_R[\tilde R_z,\rho] = -\bar\Gamma_0[\tilde R_z,\tilde R_z\rho] - \bar\Gamma_+[\tilde R^+,\tilde R^-\rho] - \bar\Gamma_-[\tilde R^-,\tilde R^+\rho] + \text{H.c.} \quad (12)$$

Here, $\bar\Gamma_0 = \gamma(\omega_L)\sin^2 2\theta\cos^2 2\phi/4 + \sin^2 2\phi[\gamma(\omega_L+\omega)\cos^4\theta + \gamma(\omega_L-\omega)\sin^4\theta]/4$, $\bar\Gamma_+ = \gamma(\omega_L+2\bar G_R)\sin^2 2\phi\sin^2 2\theta/4 + \gamma(\omega_L+\omega+2\bar G_R)\cos^4\phi\cos^4\theta + \gamma(\omega_L-\omega+2\bar G_R)\sin^4\theta\sin^4\phi$, and $\bar\Gamma_- = \gamma(\omega_L-2\bar G_R)\sin^2 2\phi\sin^2 2\theta/4 + \gamma(\omega_L+\omega-2\bar G_R)\cos^4\theta\sin^4\phi + \gamma(\omega_L-\omega-2\bar G_R)\sin^4\theta\cos^4\phi$ with $\cot 2\phi = \bar\Delta/\bar G$, and $\bar G_R = \sqrt{\bar\Delta^2 + \bar G^2}$. The new operators, i.e., $\tilde R^+ = |\tilde 2\rangle\langle\tilde 1|$, $\tilde R^- = [\tilde R^+]^\dagger$, and $\tilde R_z = |\tilde 2\rangle\langle\tilde 2| - |\tilde 1\rangle\langle\tilde 1|$, are operating in the double dressed-state picture, obeying the following commutation relations: $[\tilde R^+,\tilde R^-] = \tilde R_z$ and $[\tilde R_z,\tilde R^\pm] = \pm 2\tilde R^\pm$. The master equation (12) contains only slowly varying terms in the spontaneous emission damping, that is, we have assumed that $\bar G_R \gg \gamma(\tilde\omega)$, with $\gamma(\tilde\omega) = 2d^2\tilde\omega^3/(3\hbar c^3)$ being the single-molecule spontaneous decay rate corresponding to the double dressed-state frequency $\tilde\omega$ (see Fig. 1).

The steady-state solution of Eq. (12) can be chosen in the form

$$\rho = Z^{-1}\exp[-\eta\tilde R_z], \quad (13)$$

where the normalization Z is determined from the relation $\mathrm{Tr}\{\rho\} = 1$. Inserting (13) into (12), one obtains $\eta = \ln(\bar\Gamma_+/\bar\Gamma_-)/2$. Using the relations (7) and (11), we arrive at the following expression for the mean value of the bare-state inversion operator $\langle S_z\rangle$ represented via the double dressed-state inversion operator $\langle\tilde R_z\rangle$, respectively:

$$\langle S_z\rangle = \cos[2(\theta - \phi)]\langle\tilde R_z\rangle/2. \quad (14)$$

The steady-state expression for the double dressed-state inversion operator can be obtained with the help of Eq. (13) and the coherent molecular state $|s\rangle \equiv |N-s,s\rangle$, which denotes a symmetrized $N-$ molecule state in which $N-s$ molecules are in the lower double dressed-state $|\tilde 1\rangle$ and s molecules are excited to the upper double dressed-state $|\tilde 2\rangle$, respectively [27]. Thus

$$\langle\tilde R_z\rangle = -N + \frac{(x^{1+N}-1) - (1+N)(x-1)}{2^{-1}(x-1)(x^{1+N}-1)}, \quad (15)$$

where $x = \bar\Gamma_+/\bar\Gamma_-$. In this case the molecular system has to occupy a volume with linear dimensions of the order of the smallest relevant emission wavelength or less. However, this restriction may be relaxed for certain geometries [25–28]. The molecular operators entering in Eqs. (12)–(15) are then collective ones, i.e., $\tilde{R}^{\pm} = \sum_{j=1}^{N} \tilde{R}_j^{\pm}$ as well as $\tilde{R}_z = \sum_{j=1}^{N} \tilde{R}_{zj}$. Figure 2(b) shows the bare-state inversion operator $\langle S_z \rangle / N$, based on the expressions (14) and (15), for a collection of $N = 50$ molecules in a volume $(2\pi c/\omega_{21})^3$ or a molecular density $\bar{N} \approx 7.8 \times 10^{14}$ cm^{-3}. Almost full inversion in the steady state is achieved when we consider that the double-dressed decay rates $\gamma(\tilde{\omega})$ are equal and denoted by γ. Furthermore, abrupt population behaviors are observed as well which may be used for engineering optical switching devices with switching times of the order of $(N\gamma)^{-1}$. Note that the mean values of nondiagonal terms resulting from Eq. (12) are zero in the steady state.

We proceed by calculating the refractive properties of a very weak field probing the strongly driven molecular sample around the first laser's frequency ω_L. The linear susceptibility $\chi(\nu)$ of the probe field, at frequency ν, can be represented in terms of the Fourier transform of the average value of the two-time commutator of the molecular operator as

$$\chi(\nu) = \frac{i}{\hbar} \bar{N} d^2 \int_0^{\infty} d\tau e^{i(\nu - \omega_L)\tau} \langle [S^-(\tau), S^+] \rangle_s. \qquad (16)$$

Note that the subindex s means steady state. Inserting the dressed-state transformations (3) and (11) in (16), in a frame rotating also at the second laser's frequency ω, and using the master equation (12), one arrives at the following expression for the susceptibility:

$$\begin{aligned} \chi(\nu) = \frac{i}{\hbar} \bar{N} d^2 \langle \tilde{R}_z \rangle \{ &(1/4) \sin^2 2\theta \sin^2 2\phi [\chi_1(\Delta_p, 2\bar{G}_R) \\ &- \chi_1(\Delta_p, -2\bar{G}_R)] + \cos^4\theta [\sin^4\phi\, \chi_2(\Delta_p, 2\bar{G}_R, -\omega) \\ &- \cos^4\phi\, \chi_2(\Delta_p, -2\bar{G}_R, -\omega)] + \sin^4\theta [\cos^4\phi \\ &\times \chi_3(\Delta_p, 2\bar{G}_R, \omega) - \sin^4\phi\, \chi_3(\Delta_p, -2\bar{G}_R, \omega)] \}, \quad (17) \end{aligned}$$

where $\chi_1(\Delta_p, x) = [\bar{\Gamma}_s + i(\Delta_p + x)]/[\bar{\Gamma}_s^2 + (\Delta_p + x)^2]$, $\chi_2(\Delta_p, x, -y) = [\bar{\Gamma}_s + i(\Delta_p + x - y)]/[\bar{\Gamma}_s^2 + (\Delta_p + x - y)^2]$, and $\chi_3(\Delta_p, x, y) = [\bar{\Gamma}_s + i(\Delta_p + x + y)]/[\bar{\Gamma}_s^2 + (\Delta_p + x + y)^2]$, whereas $\Delta_p = \nu - \omega_L$ while $\bar{\Gamma}_s = 4\bar{\Gamma}_0 + \bar{\Gamma}_+ + \bar{\Gamma}_-$. Figure 4 shows the steady-state behavior of the linear susceptibility when the molecular sample is probed with a weak coherent field of frequency ν. Both positive or negative dispersions without absorption are clearly visible around $\Delta_p/\gamma = 0$, which may lead to enhanced or reduced refractive indices applicable for optical imaging, lithography, or negative refraction processes in dense media [29]. In particular, the index of refraction close to vanishing absorption $n(\nu) \approx \sqrt{1 + \chi'}$ takes values $n > 2$ for $\bar{N} = 10^{17}$ cm^{-3} and $\Delta/(2\Omega) = -0.43$. These dependencies for the susceptibility $\chi(\nu)$ are easily understood in the double dressed-state picture [see Fig. 1(c)]. Particularly, for $\Delta/(2\Omega) = 0.43$ there are more molecules in the upper double dressed state $|\tilde{2}\rangle$, i.e., $\langle \tilde{R}_z \rangle > 0$ [see Fig. 3(b)]. As a consequence, at frequency $\omega_L + \omega + 2\bar{G}_R$ one has gain, while at frequency $\omega_L - \omega - 2\bar{G}_R$ we have absorption. [See the lateral dip and peak in Fig. 4(a).] Similarly, one can explain the whole structure shown in Figs. 4(a) and 4(b). An interesting issue here is the structure at $\omega_L \pm 2\bar{G}_R$, which may help to extract the value of permanent dipoles, i.e., $|d_{22} - d_{11}|$. If one inspects the absorption spectrum shown in Fig. 4, then the frequency separation between the first maximum and minimum around $\Delta_p = 0$ is equal to $4\bar{G}_R$. $\bar{G}_R$ involves the difference of permanent dipoles $|d_{22} - d_{11}|$. This, in principle, allows one to determine $|d_{22} - d_{11}|$ if all other involved parameters are known. Finally, the elastic photon scattering spectrum consists of three lines at $\{\omega_L, \omega_L \pm \omega\}$. The inelastic one may contain up to nine spectral lines, in strict concordance with the double dressed-state formalism schematically shown in Fig. 1(c). Suppression of a spectral line at the frequency of the strongly driving laser also occurs [30], and the emission spectrum permits determination of the diagonal dipole matrix elements (see also Ref. [31]). The magnitudes of permanent dipoles are required for, e.g., interpretation of biological images, as well as for biological high-harmonic or ultrashort pulse generation processes via laser-pumped media possessing permanent dipoles [32] (see also [33]).

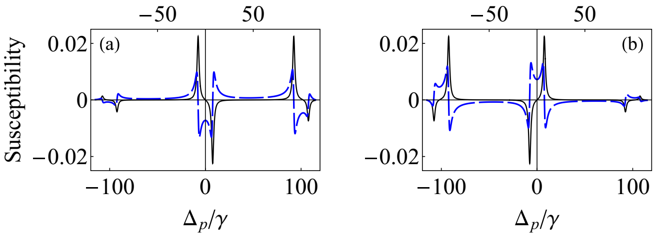

FIG. 4. (Color online) The steady-state dependence of the linear susceptibility $\chi(\nu_p)$ [in units of $\bar{N} d^2/(\hbar\gamma)$] versus scaled detuning Δ_p/γ. The solid black curve corresponds to the imaginary part (absorption spectrum) and the long-dashed blue line to the real part of the susceptibility, respectively. (a) $\Delta/(2\Omega) = 0.43$ and (b) $\Delta/(2\Omega) = -0.43$. Other parameters are $\Omega/\gamma = 45$, $\omega/\gamma = 100$, and $G/\gamma = 16$.

IV. SUMMARY

Summarizing, we have investigated the steady-state quantum dynamics of laser-pumped two-level molecular samples with broken inversion symmetry. We have demonstrated population inversion in the bare states due to induced coherences which in turn depend on the magnitude of permanent dipoles. Vacuum-mediated collective effects among the two-level emitters considerably enhance the molecular inversion. The values of the permanent dipoles can be inferred from the emission or absorption spectra. Furthermore, the investigated system exhibits large positive or negative dispersion without absorption, facilitating applications including optical imaging, lithography, and negative indices of refraction. The results apply especially to biomolecular, spin, or asymmetrical quantum dot systems.

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