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Tunable ultra-fast carrier–light field dynamics of quantum dots

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ABSTRACT A theory of ultra-fast carrier–light field dynamics of quantum dots is presented. The carrier–light field dynamics is described by Maxwell–Bloch equations. A calculation of the dipole matrix elements requires the determination of the electronic wave functions taking into account their dependence on the degeneracy of the carrier states. The ultra-fast carrier–light field dynamics depends strongly on the external applied electric field.

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1 Introduction

The nano-optics of self-organized zero-dimensional semiconductor structures (quantum dots, QDs) [1–3] has recently attracted increased attention for both fundamental and application-related reasons (for a recent review of the field see e.g. [4]). In particular, with the dependence of their optoelectronic properties on size and shape the possibility of employing quantum dots as ‘tunable’ active media makes them most attractive for long-wavelength semiconductor lasers. Most prominently, the opto-electronic properties of quantum dots are reflected in their discrete multilevel energy spectrum. The level system depends strongly on the size of the quantum dots [5] and the carriers. The carrier dynamics is characterized by their degeneracy due to the coupling of states in the different quantization directions. The dynamics also depends on the anisotropy concerning arbitrary shapes of the carrier-confining potentials, the number of the states as well as the carrier–carrier, carrier–phonon and carrier–field many-body interactions.

In this article, we sketch a theory for the optics, in the presence and absence of an external static electric field, of an ensemble of quantum dots which are coupled only by an optical field (but not by other mechanisms). It may be regarded as a basis for an investigation of the dynamics in terms of suitable Maxwell–Bloch equations [6, 7]. Here, we concentrate on a system of cubic quantum dots, made with GaAs embedded in the background material AlGaAs. Although the

presence of an external electric or magnetic field in quantum dots has attracted the interests of experimental and theoretical groups [4], in this paper we will focus on the theoretical aspects of the application of an external electric field and not on the experimental overviews.

2 Theoretical description

We describe the theory of the interaction of an optical field with the carriers confined in quantum dots. To properly take into account the three-dimensional confinement of the carriers within the quantum dots, we formulate the theory in terms of the actual number of carriers within each discrete energy level. The resulting Hamiltonian for the quantum dots comprises contributions of the free carriers (within the effective-mass approximation) and the coupling of the light field with the carriers. Here, we will concentrate on the light-field coupling. For simplicity we will disregard band-mixing effects. We introduce the carrier position vector inside the QD $\mathbf{r}' = (x', y', z')$ and vectorial indices $\mathbf{i} = (i_{x'}, i_{y'}, i_{z'})$, $\mathbf{j} = (j_{x'}, j_{y'}, j_{z'})$ for electrons and holes, respectively, where the subindices denote the directions of the quantization to describe the degeneracy of the carrier energy level spectrum. The wavefunctions $\Psi_{i(j)}^{e(h)}(\mathbf{r}')$ are determined by the Schrödinger equation for electrons and holes:

$$\left[-\frac{\hbar^2}{2m^{*e(h)}} \nabla^2 + V^{e(h)}(\mathbf{r}') - e\mathbf{r}' \cdot \mathcal{E}(\mathbf{r}') \right] \Psi_{i(j)}^{e(h)}(\mathbf{r}') = \varepsilon_{i(j)}^{e(h)} \Psi_{i(j)}^{e(h)}(\mathbf{r}'), \quad (1)$$

where $m^{*e(h)}$ is the effective mass for electrons (holes) and $\mathcal{E}(\mathbf{r}')$ is an external static electric field. The wavefunctions $\Psi_{i(j)}^{e(h)}(\mathbf{r}')$, the confining potential $V_0(\mathbf{r}')$ and the carrier energies $\varepsilon_{i(j)}^{e(h)}$ are given by $\Psi_i^e(\mathbf{r}') = \Psi_{i_{x'}}^e(x') \Psi_{i_{y'}}^e(y') \Psi_{i_{z'}}^e(z')$ (similarly for holes), $V^{e(h)}(\mathbf{r}') = V_0^{e(h)} \Theta\left(\frac{L_x}{2} - |x|\right) \Theta\left(\frac{L_y}{2} - |y|\right) \Theta\left(\frac{L_z}{2} - |z|\right)$ (Θ is the Heaviside step function) and $\varepsilon_i^e = \varepsilon_{i_{x'}} + \varepsilon_{i_{y'}} + \varepsilon_{i_{z'}}$ (similarly for holes). For the calculation of the carrier wavefunctions and the formulation of the Hamiltonian contributions we assume cubic confining potentials. As has recently been shown, the electronic properties of QDs are essentially independent of the dot shape [8]; however, they are strongly dependent on

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their size. The three-dimensional carrier confinement introduces degeneracy of the carrier energy spectrum. As a result, the total carrier energy is obtained by summation of the carrier energies in each quantized direction. For determination of the macroscopic polarization, which describes the strength of the internal field induced by the optical field, the dipole matrix elements $\mathbf{d}_{cv}^{i,j}$ have to be determined. In terms of vectorial indices they read $\mathbf{d}_{cv}^{i,j} = \gamma \int \Psi^e(\mathbf{r}') \Psi^h(\mathbf{r}') d\mathbf{r}'$, where $\gamma = (\hbar e / im^*) \int_{V_E} v_k^e v_k^h \mathbf{r}' d\mathbf{r}'$ is the coupling factor [9], V_E is the volume of the unit cell and v_k^e (v_k^h) denote the Bloch functions for electrons (holes). On a semiclassical basis the carrier–light field interaction Hamiltonian is given by

$$H_{C-L}(\mathbf{r}, t) = - \sum_{i,j} \left[\mathbf{d}_{cv}^{i,j} \cdot \mathbf{E}^-(\mathbf{r}, t) \hat{c}_i^\dagger \hat{d}_j^\dagger + \mathbf{d}_{cv}^{i,j} \cdot \mathbf{E}^+(\mathbf{r}, t) \hat{d}_j \hat{c}_i \right], \quad (2)$$

where \hat{c}_i and \hat{c}_i^\dagger (\hat{d}_j and \hat{d}_j^\dagger) denote the annihilation and creation operators, respectively, for electrons (holes). $\mathbf{E}^+(\mathbf{r}, t)$ and $\mathbf{E}^-(\mathbf{r}, t)$ are the positive and negative frequency components [7] of the classical optical field $\mathbf{E}(\mathbf{r}, t)$, respectively. The Hamiltonian is given in the rotating wave approximation.

The interplay of the optical light field and the matter is described by the Maxwell–Bloch equations. They consist of wave equations for the propagating light field and Bloch equations for the spatial–temporal dynamics of the active charge carrier plasma within the quantum-dot medium. The coupled system of partial differential equations then constitutes the quantum-dot Maxwell–Bloch equations that model on a mesoscopic basis the dynamic light–matter interactions occurring within a QD laser. Via the polarization of the active QD medium, the light field is locally coupled to the dynamics of the carriers and to the inter-level dipole dynamics (described on the basis of the QD Bloch equations). Thereby, the individual time scales of the underlying optical and electronic processes spanning a temporal regime from femtoseconds up to nanoseconds are self-consistently included. The dynamics of the occupation of electrons (e, level index ‘ i ’) and holes (h, level index ‘ j ’), $n_i^{e,h}$, and the dynamics of the inter-level polarizations p^\pm within a QD, are governed by the QD Bloch equations (where \mathbf{r} denotes the positions of the QDs in the array)

$$\begin{aligned} n_i^e &= \langle \hat{c}_i^\dagger \hat{c}_i \rangle, & p_{i,j}^+ &= \langle \hat{c}_i^\dagger \hat{d}_j^\dagger \rangle, \\ n_j^h &= \langle \hat{d}_j^\dagger \hat{d}_j \rangle, & p_{i,j}^- &= \langle \hat{d}_j \hat{c}_i \rangle, \end{aligned} \quad (3)$$

$$\begin{aligned} \frac{\partial}{\partial t} n_i^e(\mathbf{r}, t) &= \Lambda_i^e(\mathbf{r}, t) - d_i^e(\mathbf{r}, t) - \gamma_i^e \left[n_i^e(\mathbf{r}, t) - n_{eq,i}^e(\mathbf{r}, t) \right] \\ &\quad - \sum_j \gamma_{i,j}^{sp} n_i^e(\mathbf{r}, t) n_j^h(\mathbf{r}, t) - \gamma_i^{nr} n_i^e(\mathbf{r}, t), \end{aligned} \quad (4)$$

$$\begin{aligned} \frac{\partial}{\partial t} n_j^h(\mathbf{r}, t) &= \Lambda_j^h(\mathbf{r}, t) - d_j^h(\mathbf{r}, t) - \gamma_j^h \left[n_j^h(\mathbf{r}, t) - n_{eq,j}^h(\mathbf{r}, t) \right] \\ &\quad - \sum_i \gamma_{i,j}^{sp} n_i^e(\mathbf{r}, t) n_j^h(\mathbf{r}, t) - \gamma_j^{nr} n_j^h(\mathbf{r}, t) \end{aligned} \quad (5)$$

and

$$\begin{aligned} \frac{\partial}{\partial t} p_{i,j}^\pm(\mathbf{r}, t) &= - \left[\gamma_{i,j}^p \mp i\tilde{\omega}_{i,j} \right] p_{i,j}^\pm(\mathbf{r}, t) \\ &\quad \mp \frac{i}{\hbar} \mathbf{d}_{cv}^{i,j} \cdot \mathbf{E}^\pm(\mathbf{r}, t) (1 - n_i^e(\mathbf{r}, t) - n_j^h(\mathbf{r}, t)), \end{aligned} \quad (6)$$

where

$$\begin{aligned} d_{i(j)}^{e(h)}(\mathbf{r}, t) &= \\ &= (i/\hbar) \sum_{i(j)} \left[\mathbf{d}_{cv}^{i,j} \cdot \mathbf{E}^-(\mathbf{r}, t) p_{i,j}^+(\mathbf{r}, t) - \mathbf{d}_{cv}^{i,j} \cdot \mathbf{E}^+(\mathbf{r}, t) p_{i,j}^-(\mathbf{r}, t) \right], \end{aligned}$$

the free rotating frequency $\tilde{\omega}_{i,j} = (1/\hbar)(\varepsilon_i^e + \varepsilon_j^h \varepsilon_{gap} - \hbar\omega)$ and ε_{gap} is the energy gap. The Bloch equations for the carrier dynamics consist of contributions describing the electrical injection of carriers (pumping) $\Lambda_{i(j)}^{e(h)}(\mathbf{r}, t)$ (including Pauli blocking), stimulated emission (with generation rate $d_{i(j)}^{e(h)}(\mathbf{r}, t)$), spontaneous recombination of the carriers $\gamma_{i,j}^{sp}$, non-radiative scattering rates $\gamma_{i(j)}^{nr}$, the Fermi distribution $n_{eq,i(j)}^{e(h)}(\mathbf{r}, t)$ and carrier-scattering rates $\gamma_{i(j)}^{e(h)}$. The dynamics of the inter-level dipole depends on the energy difference of the electron–hole pair, light frequency ω , dipole dephasing $\gamma_{i,j}^p$ and inversion.

The dynamics is mainly driven by the exchange of energy between the semiconductor and the electrical field, which leads to damped relaxation oscillations. In order to bring in the relevant time scales (frequency ω_{rel} and damping rate γ_{rel} of the relaxation oscillations) in connection with the coefficients given in (3)–(6) we give a short review of the fundamental steps in deriving the formula for the relaxation oscillations. As we stated, the source of the relaxation oscillations is the exchange of energy between the semiconductor and the electrical field, so the dynamics of the polarization itself is not directly of interest. Since the dephasing time $t_{i,j}^p = 1/\gamma_{i,j}^p \approx 500$ fs in semiconductors is normally very short, the dynamics of the polarization sticks to the temporal behavior of the field. This is expressed by an adiabatic elimination of the polarization in (6), resulting in

$$\begin{aligned} p_{i,j}^\pm(\mathbf{r}, t) &= \mp \frac{i}{\hbar} \mathbf{d}_{cv}^{i,j} \cdot \mathbf{E}^\pm(\mathbf{r}, t) (1 - n_i^e(\mathbf{r}, t) - n_j^h(\mathbf{r}, t)) \\ &\quad \times \left(\gamma_{i,j}^p \mp i\tilde{\omega}_{i,j} \right), \\ d_{i(j)}^{e(h)}(\mathbf{r}, t) &= \frac{1}{\hbar^2} \sum_{j(i)} (\mathbf{d}_{cv}^{i,j} \cdot \mathbf{E}^+(\mathbf{r}, t)) (\mathbf{d}_{cv}^{i,j} \cdot \mathbf{E}^-(\mathbf{r}, t)) \\ &\quad \times \frac{2}{\gamma_{i,j}^p} \mathcal{L}(\gamma_{i,j}^p, \tilde{\omega}_{i,j}) (n_i^e(\mathbf{r}, t) + n_j^h(\mathbf{r}, t) - 1), \\ \mathcal{L}(\gamma_{i,j}^p, \tilde{\omega}_{i,j}) &= \frac{(\gamma_{i,j}^p)^2}{(\gamma_{i,j}^p)^2 + (\tilde{\omega}_{i,j})^2}. \end{aligned} \quad (7)$$

Since the electrical field couples to the macroscopic polarization $\mathbf{P}(\mathbf{r}, t) = (1/V) \sum_{\pm,i,j} \mathbf{d}_{cv}^{i,j} p^\pm(\mathbf{r}, t)$, we also introduce the macroscopic densities $N^{e(h)}(\mathbf{r}, t) = (1/V) \sum_{i(j)} n_{i,j}^{e(h)}(\mathbf{r}, t)$. Assuming charge neutrality, i.e. $N^e(\mathbf{r}, t) = N^h(\mathbf{r}, t) = N(\mathbf{r}, t)$, we have the following equation of motion for $N(\mathbf{r}, t)$:

$$\begin{aligned} \frac{\partial}{\partial t} N(\mathbf{r}, t) &= \Lambda(\mathbf{r}, t) - D(\mathbf{r}, t) I(\mathbf{r}, t) - D^{sp}(\mathbf{r}, t) - D^{nr}(\mathbf{r}, t), \\ D(\mathbf{r}, t) &= \frac{1}{V} \sum_i X_i^e(\mathbf{r}, t) = \frac{1}{V} \sum_j X_j^h(\mathbf{r}, t), \end{aligned} \quad (8)$$

$$D^{\text{sp}}(\mathbf{r}, t) = \frac{1}{V} \sum_{i,j} \gamma_{i,j}^{\text{sp}} n_i^e(\mathbf{r}, t) n_j^h(\mathbf{r}, t),$$

$$X_{i(j)}^{e(h)}(\mathbf{r}, t) = \frac{1}{\hbar} \sum_{i(j)} |\mathbf{d}_{ij}^{\text{cv}}|^2 \cos^2 \theta_{i,j}(\mathbf{r}, t) \frac{2}{\gamma_{i,j}^{\text{p}}} \mathcal{L} \left(\gamma_{i,j}^{\text{p}}, \tilde{\omega}_{i,j} \right) \\ \times (n_i^e(\mathbf{r}, t) + n_j^h(\mathbf{r}, t) - 1),$$

with similar definitions for $\Lambda(\mathbf{r}, t)$ and $D^{\text{nr}}(\mathbf{r}, t)$. $\theta_{i,j}(\mathbf{r}, t)$ is the angle between the optical field and the dipole and the intensity $I(\mathbf{r}, t) = \mathbf{E}^+(\mathbf{r}, t) \cdot \mathbf{E}^-(\mathbf{r}, t)$.

Since it is sufficient to have a local description of the variables $N(\mathbf{r}, t)$ and $\mathbf{E}(\mathbf{r}, t)$ for an explanation of relaxation oscillations, we drop the space dependence and consider these space-independent variables as averaged variables over the extension of the laser array. If such an averaging is carried out for the electrical field in the slowly varying amplitude approximation, the time dynamics for the intensity $I(t) = (1/V) \int I(\mathbf{r}, t) d\mathbf{r}$ is given by

$$\frac{\partial}{\partial t} I(t) = (G(t) - \kappa) I(t), \quad G(t) = \frac{1}{4\beta} \hbar \omega D(t), \quad \beta = \frac{1}{2} \varepsilon_0 n_{\text{eff}}^2, \quad (9)$$

where $D(t)$ is defined by (8) and κ is the rate of cavity loss.

Linear perturbation theory around the steady-state solutions N_{ss} and I_{ss} results in the following matrix equation for the perturbation δN and δI :

$$\frac{\partial}{\partial t} \begin{pmatrix} \delta N \\ \delta I \end{pmatrix} = \begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix} \begin{pmatrix} \delta N \\ \delta I \end{pmatrix}, \quad (10)$$

$$a_{11} = -\left. \frac{\partial D}{\partial N} \right|_{N_{\text{ss}}} I_{\text{ss}} - \left. \frac{\partial D^{\text{sp}}}{\partial N} \right|_{N_{\text{ss}}} - \left. \frac{\partial D^{\text{nr}}}{\partial N} \right|_{N_{\text{ss}}}, \quad a_{12} = -D_{\text{ss}},$$

$$a_{21} = \left. \frac{\partial G}{\partial N} \right|_{N_{\text{ss}}} I_{\text{ss}}, \quad a_{22} = G_{\text{ss}} - \kappa = 0.$$

For (10) we have the following eigenvalues λ :

$$\lambda_{1,2} = \frac{1}{2} \left[a_{11} \pm \sqrt{a_{11}^2 + 4a_{12}a_{21}} \right] = \gamma_{\text{rel}} \pm i\omega_{\text{rel}}, \quad (11)$$

$$\text{where } \omega_{\text{rel}} = \sqrt{|\gamma_{\text{rel}}^2 + a_{12}a_{21}|}. \quad (12)$$

3 Numerical results and conclusions

In Fig. 1a are presented the first electron and hole energy levels for a single QD made with GaAs/Al_{0.2}Ga_{0.8}As as a function of external electric field and in Fig. 1b the energy difference between the energy summation of the carriers and the energy summation without an electric field. In Fig. 1c is presented the dipole matrix element value versus the electric field. Obviously, the carrier energies become larger by applying an electric field. This enlargement influences the free rotating frequency. (The material parameters for GaAs/Al_{0.2}Ga_{0.8}As have been taken from Adachi [10].) In Fig. 2 are illustrated the intensity (power) and density as a function of time in the absence (Fig. 2a) and presence (Fig. 2b) of an external electric field for a sample of uncoupled QDs (10 × 10 × 10 nm). We consider only the ground state of electrons and holes for the present calculations. From

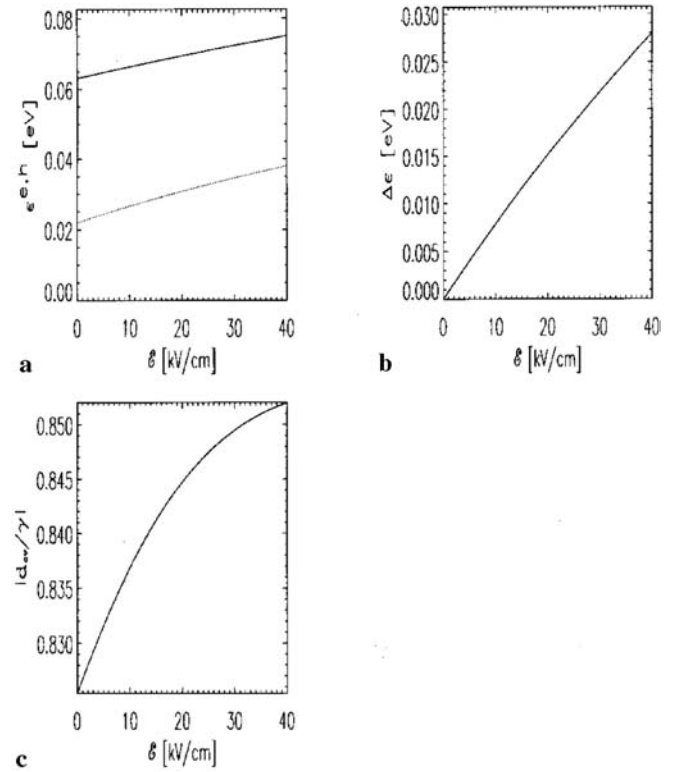


FIGURE 1 **a** The first energy levels for an electron (*solid line*) and a hole (*dotted line*) for a QD (10 × 10 × 10 nm) as a function of external electric field. **b** The energy difference between electron–hole summation and the energy summation corresponding to zero electric field. **c** Normalized dipole matrix element as a function of external electric field with the same geometry as for (a)

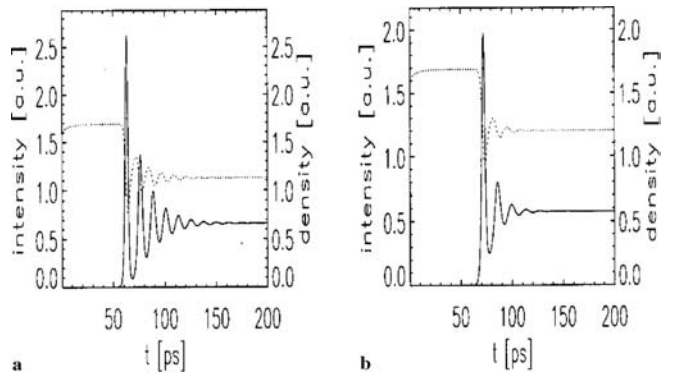


FIGURE 2 **a** Intensity (*solid line*) and density (*dotted line*) for QDs (10 × 10 × 10 nm) in the absence of an external electric field. **b** Intensity (*solid line*) and density (*dotted line*) for QDs (10 × 10 × 10 nm) in the presence of $\mathcal{E}(\mathbf{r}') = \mathcal{E}_z' = 7 \times 10^3$ V/cm

(12) we get the following values for the damping rates and the frequency of the relaxation oscillations: $\gamma_{\text{rel}} = 0.07 \text{ ps}^{-1}$, $\omega_{\text{rel}} = 0.48 \text{ ps}^{-1}$ without external electric field (Fig. 2a) and $\gamma_{\text{rel}} = 0.11 \text{ ps}^{-1}$, $\omega_{\text{rel}} = 0.45 \text{ ps}^{-1}$ with external electric field (Fig. 2b). These values are quite close to those which emerge from Fig. 2. The application of an external electric field is accompanied by a change in the carrier energies and the dipole matrix elements. The carrier energies directly influence the value of the free rotating frequency $\tilde{\omega}_{i,j}$ and hence the value of the Lorentzian \mathcal{L} . This is far more sensitive than a direct change in the coefficients due to a different dipole matrix

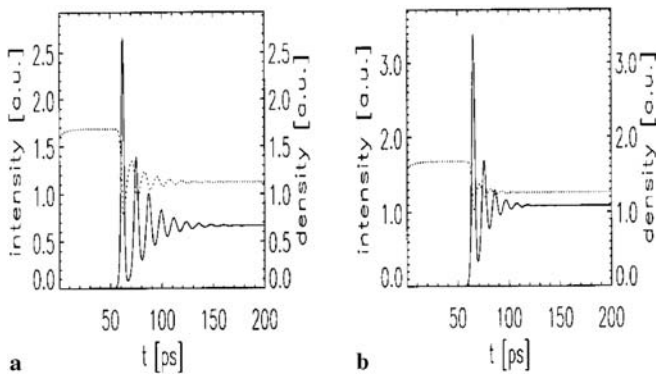


FIGURE 3 **a** Intensity (solid line) and density (dotted line) for QDs ($10 \times 10 \times 10$ nm) in the absence of an external electric field and dipole matrix element value for $\mathcal{E}(\mathbf{r}') = \mathcal{E}_{z'} = 7 \times 10^3$ V/cm. **b** Intensity (solid line) and density (dotted line) for QDs ($10 \times 10 \times 10$ nm) in the presence of $\mathcal{E}(\mathbf{r}') = \mathcal{E}_{z'} = 7 \times 10^3$ V/cm and dipole matrix element value in the absence of an external field

element. Since the damping rate $\gamma_{\text{rel}} = 2a_{11}$ is directly dependent on the Lorentzian we expect a larger change in the damping rate than a change in the frequency of relaxation oscillations, as is illustrated in Fig. 2.

The proof of this can be seen with the aid of Fig. 3. Here either the transition energy or the dipole matrix elements have been changed but not both at the same time. In Fig. 3a are illustrated the intensity and density as a function of time in the absence of an external electric field, meaning that the transition energy is not changed. The value of the dipole matrix element is that corresponding to an electric field $\mathcal{E}_{z'} = 7 \times 10^3$ V/cm. Since the change in relaxation oscillations is obviously small (compare with Fig. 2a), it is apparent that the dynamics is mainly affected by a change in transition

frequency that goes with the application of an external electric field. In Fig. 3b the transition energy is that corresponding to an electric field $\mathcal{E}_{z'} = 7 \times 10^3$ V/cm and the dipole moment has the zero-field value. A comparison with Fig. 2b reveals primarily the increased damping rate γ_{rel} , with the enlarged transition energy with electric field. For $\mathcal{E}_{z'} = 7 \times 10^3$ V/cm the change in transition energy $\Delta\varepsilon$ from Fig. 1b is 5 meV, which results in a change $\Delta\tilde{\omega} = 7.5 \text{ ps}^{-1}$. This value has the same order of magnitude as $\gamma^{\text{p}} = 2 \text{ ps}^{-1}$ despite the relatively small change in transition energy. The denominator of the Lorentzian changes considerably.

The possibility of increasing the transition energy with the strength of the electric field introduces the opportunity to control the mismatch of the resonator wavelength and the spectral position of the gain due to a decreased band-gap energy at higher lattice temperatures. This limits the maximum attainable power of such devices.

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