Collective Excitations and the Dynamical Stark Effect in a Coherently Driven Exciton System

S. Schmitt-Rink
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

and

D. S. Chemla
AT&T Bell Laboratories, Holmdel, New Jersey 07733
(Received 8 September 1986)

We consider a semiconductor of arbitrary dimension subject to a strong monochromatic laser beam in the transparency region below the exciton resonance. We calculate the spectrum of collective excitations and explain the recently observed dynamical Stark effect of the exciton. For the first time, we derive from first principles the nonresonant nonlinear susceptibility $\chi^{(3)}$ including exciton correlations.

PACS numbers: 71.35.+z, 71.45.Gm, 78.45.+h

The nonlinear optical properties of excitons in semiconductors are of wide current interest, both from a fundamental physics as well as from a device point of view. Recent time-resolved measurements of the excitonic optical absorption under pumping *below* the absorption edge revealed a new and interesting phenomenon, an ultrafast dynamical blue shift of the exciton resonance accompanied by a strong bleaching.[1,2] The purpose of this paper is to explain this "ac Stark effect," which is of great technological importance because of its potential applications in ultrafast optical signal processing, and to derive from first principles the effects of exciton correlations on the nonresonant nonlinear susceptibility $\chi^{(3)}$.

The ac Stark effect has been extensively studied in atomic vapors and was also observed in the yellow series of Cu_2O under pumping of the $1s$-to-$2p$ exciton transition.[3] In both cases, the essential physics is that of a two-level system "dressed" by the pump photons and the effect is easily analyzed.[4] In the present case, the effect occurs under nonresonant pumping of the ground-state to exciton transition and is therefore *quite different in character.* The essential point is that excitons are *not* ideal Bose particles (with polaritons being the "dressed" particles), but composite particles made from electrons and holes. It is exactly this internal structure that gives rise to residual (anharmonic) interactions, with both the light field and other excitons (electrons and holes), which ultimately determine almost all excitonic optical nonlinearities,[5] including the present one. Since the absorption tail in semiconductors is negligibly (exponentially) small, *no real exciton population,* and thus no lifetime, is involved in the ac Stark effect. (The situation is thus different from that in atomic vapors, where the absorption tail is a Lorentzian.) The effect is solely induced by the electric field of the nonresonant pump beam and persists for its duration only. Below, we show that the virtual excitons created give rise to exactly the same physical processes as real ones, which allows for an intuitive physical interpretation of all the experimental observations.

We begin by considering a semiconductor in the presence of a strong monochromatic pump beam $E_p^* \times\exp(i\omega_p t)+E_p\exp(-i\omega_p t)$ in the transparency region below the exciton resonance. Within the rotating frame, we characterize it by a matrix distribution function

$$\hat{n}_{\mathbf{k}}=\begin{pmatrix}\langle c_{1\mathbf{k}}^{\dagger}c_{1\mathbf{k}}\rangle & \langle c_{1\mathbf{k}}^{\dagger}c_{2\mathbf{k}}\rangle\\ \langle c_{2\mathbf{k}}^{\dagger}c_{1\mathbf{k}}\rangle & \langle c_{2\mathbf{k}}^{\dagger}c_{2\mathbf{k}}\rangle\end{pmatrix}=\begin{pmatrix}n_{1\mathbf{k}} & \psi_{\mathbf{k}}^{*}\\ \psi_{\mathbf{k}} & n_{2\mathbf{k}}\end{pmatrix} \tag{1}$$

and an energy matrix

$$\hat{\varepsilon}_{\mathbf{k}}=\begin{pmatrix}\varepsilon_{1\mathbf{k}}^{0} & -\mu^{*}E_p^{*}\\ -\mu E_p & \varepsilon_{2\mathbf{k}}^{0}\end{pmatrix}-\sum_{\mathbf{k}'}V_{\mathbf{k},\mathbf{k}'}\hat{n}_{\mathbf{k}'}, \tag{2}$$

subject to $i\,\partial\hat{n}_{\mathbf{k}}/\partial t=[\hat{n}_{\mathbf{k}},\hat{\varepsilon}_{\mathbf{k}}]=0$. 1 and 2 denote the conduction (c) and valence (v) bands with dispersions $\varepsilon_{1\mathbf{k}}^{0}=(E_g-\omega_p)/2+k^2/2m_e$ and

$$\varepsilon_{2\mathbf{k}}^{0}=-(E_g-\omega_p)/2-k^2/2m_h+\sum_{\mathbf{k}'}V_{\mathbf{k},\mathbf{k}'},$$

respectively; E_g is the band gap, $\mu=er_{cv}$ the interband dipole matrix element (assumed to be constant), and $V_{\mathbf{k},\mathbf{k}'}$ the Coulomb interaction. Spin indices are suppressed.

Here, we have assumed that (i) the coherent pump field can be treated classically, (ii) the rotating-wave approximation holds, (iii) relaxation processes can be neglected, and (iv) many-body effects (including electron-hole correlation) can be treated within the Hartree-Fock (HF) approximation, giving rise to a "molecular field" $-\sum_{\mathbf{k}'}V_{\mathbf{k},\mathbf{k}'}\hat{n}_{\mathbf{k}'}$. Approximations (i), (ii) and (iii) are justified for the experiments of interest. As for the rather crude approximation (iv), we already note that our final results can be easily generalized.

The eigenvalues of the energy matrix (2) yield the quasiparticle ["dressed" electrons (e) and holes (h)] dispersion in the rotating frame:

$$e_{\mathbf{k}}^{\pm} = \tfrac{1}{2}\{\varepsilon_{1\mathbf{k}} + \varepsilon_{2\mathbf{k}} \pm [(\varepsilon_{1\mathbf{k}} - \varepsilon_{2\mathbf{k}})^2 + 4|\Delta_{\mathbf{k}}|^2]^{1/2}\}, \quad (3)$$

where

$$\Delta_{\mathbf{k}} = \mu E_p + \textstyle\sum_{\mathbf{k}'} V_{\mathbf{k},\mathbf{k}'} \psi_{\mathbf{k}'}$$

and

$$\varepsilon_{i\mathbf{k}} = \varepsilon_{i\mathbf{k}}^0 - \textstyle\sum_{k'} V_{\mathbf{k},\mathbf{k}'} n_{i\mathbf{k}'}, \quad i = 1,2,$$

are the renormalized (HF) electron ($\varepsilon_{1\mathbf{k}}$) and hole ($-\varepsilon_{2-\mathbf{k}}$) energies. Since $\varepsilon_{1\mathbf{k}} - \varepsilon_{2\mathbf{k}} > 0$, Eq. (3) simply describes (light-)shifted conduction and valence bands. A band splitting at ω_p occurs under resonant excitation only, in which case, however, relaxation processes must be included.[6]

From $[\hat{n}_{\mathbf{k}}, \hat{\varepsilon}_{\mathbf{k}}] = 0$, we obtain for the light-induced e-h pair amplitude $\psi_{\mathbf{k}}$

$$(\varepsilon_{1\mathbf{k}} - \varepsilon_{2\mathbf{k}})\psi_{\mathbf{k}} = (n_{2\mathbf{k}} - n_{1\mathbf{k}})(\mu E_p + \textstyle\sum_{k'} V_{\mathbf{k},\mathbf{k}'} \psi_{\mathbf{k}'}), \quad (4)$$

and for the distribution function $n_{\mathbf{k}} = 1 - n_{2\mathbf{k}} = n_{1\mathbf{k}}$

$$n_{\mathbf{k}} = \tfrac{1}{2}[1 - \mathrm{sgn}(\varepsilon_{1\mathbf{k}} - \varepsilon_{2\mathbf{k}})(1 - 4|\psi_{\mathbf{k}}|^2)^{1/2}]. \quad (5)$$

$\psi_{\mathbf{k}}$ satisfies an exciton Schrödinger equation (4), driven by the pump beam and including (self-consistent) exchange and phase-space filling (blocking) corrections, the physics of which has been discussed elsewhere.[5] The (virtual) e-h pair density is determined by $n = 2\sum_{\mathbf{k}} n_{\mathbf{k}}$ and for $n a_0^d \ll 1$, where a_0 is the exciton bohr radius, we obtain from (5) $n_{\mathbf{k}} \simeq |\psi_{\mathbf{k}}|^2$. Throughout this paper we will consider this limit of a weakly nonideal (virtual) exciton gas, in which the distribution functions are determined by the probability of finding the (virtual) electrons and holes in (unperturbed) exciton states $|\psi\rangle$. Self-energy and vertex corrections combine them simply to effective exciton-exciton interactions.

The nonlinear optical susceptibility χ_p experienced by the pump beam is obtained from the induced polarization amplitude $P_p = 2\mu^* \sum_{\mathbf{k}} \psi_{\mathbf{k}}$; $\chi_p = P_p/E_p$. Expanding the eigenfunctions of the homogeneous part of (4) in terms of the unperturbed exciton wave functions $\phi_{n\mathbf{k}}$, we obtain in leading order in the e-h pair density $\chi_p = 2\sum_n f_n^{\mathrm{H}}/(E_n^{\mathrm{H}} - \omega_p)$, where

$$f_n^{\mathrm{H}} = |\mu|^2 \sum_{\mathbf{k},\mathbf{k}'} \left[\phi_{n\mathbf{k}}(1 - 2n_{\mathbf{k}'})\phi_{n\mathbf{k}'}^* - \sum_{\substack{m \\ m \neq n}} \frac{\phi_{n\mathbf{k}} \Sigma_{nm}^{\mathrm{H}} \phi_{m\mathbf{k}'}^* + (n \leftrightarrow m)}{E_m^0 - E_n^0} \right] \quad (6)$$

are the renormalized oscillator strengths and $E_n^{\mathrm{H}} = E_n^0 + \Sigma_{nn}^{\mathrm{H}}$ the renormalized transition energies. E_n^0 are the unperturbed exciton energies and $\Sigma_{nm}^{\mathrm{H}} = \langle n\psi | I_x | m\psi \rangle$ the Hartree exciton self-energies, where I_x denotes the exciton exchange interaction [i.e., $(V_t - V_s)/2$, where V_t and V_s are the interactions in the triplet and singlet channels, respectively[5]) and $|\psi\rangle = \mu E_p \sum_n [\phi_n^*(\mathbf{r}=0)/(E_n^0 - \omega_p)]|n\rangle$. As a result of charge neutrality, the direct exciton interaction [i.e., $(V_t + V_s)/2$] does not contribute.[5]

$f_n^{\mathrm{H}}(6)$ and E_n^{H} are the generic expressions determining $\chi_p^{(3)}$, if Σ_{nm}^{H} is generalized to include the effects of multiple exciton-exciton scattering in both intermediate and final states. In any case, the (dispersive) optical nonlinearity is due to (i) a shift of the exciton energies as a result of exciton-exciton collisions, (ii) a corresponding exciton wave-function renormalization [second term in (6)], and (iii) a phase-space filling (blocking) correction to the oscillator strength [first term in (6)].

The nonresonant, coherent pump beam induces a virtual, coherent e-h pair state, similar to a (real) Bose condensed state, the "symmetry breaking" being externally imposed rather than spontaneous. A weak test beam $E_t^* \exp(i\omega_t t) + E_t \exp(-i\omega_t t)$ probes the excitation spectrum of this state, and thus the polarization induced by it has to be clearly distinguished from that induced by the pump beam.

The test beam induces changes $\delta\hat{n}_{\mathbf{k}}$ of the matrix distribution function $\hat{n}_{\mathbf{k}}$ oscillating at frequencies $\pm\Delta\omega$, where $\Delta\omega = \omega_t - \omega_p$. Different from noninteracting systems, $\delta\hat{n}_{\mathbf{k}}$ reacts in turn on the energies (2), giving rise to an induced, self-consistent potential. If we assume for the moment that the external perturbation does not conserve momentum, we obtain

$$i\frac{\partial\, \delta\hat{n}_{\mathbf{k}}(\mathbf{q})}{\partial t} = \delta\hat{n}_{\mathbf{k}}(\mathbf{q})\hat{\varepsilon}_{\mathbf{k}+\mathbf{q}} - \hat{\varepsilon}_{\mathbf{k}}\delta\hat{n}_{\mathbf{k}}(\mathbf{q}) + \hat{n}_{\mathbf{k}}\delta\hat{\varepsilon}_{\mathbf{k}}(\mathbf{q}) - \delta\hat{\varepsilon}_{\mathbf{k}}(\mathbf{q})\hat{n}_{\mathbf{k}+\mathbf{q}}, \quad (7)$$

where

$$\delta\hat{\varepsilon}_{\mathbf{k}}(\mathbf{q}) = -\hat{\tau}_+ \mu^* E_t^* e^{i\Delta\omega t} - \hat{\tau}_- \mu E_t e^{-i\Delta\omega t} - \textstyle\sum_{\mathbf{k}'} V_{\mathbf{k},\mathbf{k}'} \delta\hat{n}_{\mathbf{k}'}(\mathbf{q}) + \hat{\tau}_0 2V_q \sum_{\mathbf{k}'} \mathrm{tr}\,\delta n_{\mathbf{k}'}(\mathbf{q}) \quad (8)$$

is the effective (external plus induced) potential seen by the system. $\hat{\tau}_{\pm} = (\hat{\tau}_1 \pm i\hat{\tau}_2)/2$ are Pauli matrices and $\hat{\tau}_0$ is the unit matrix. The first three terms in (8) account for the fact that the system responds to the exact local (external plus molecular) field, and the last term in (8) describes the screening of the perturbation, the importance of which (in this

context) was first emphasized by Anderson.[7] It is exactly this last term which (because of the long-range nature of the Coulomb interaction) forces us to keep track of the $\mathbf{q}$ dependence of $\delta\hat{n}_{\mathbf{k}}(\mathbf{q})$.

Equations (7) and (8) are in the spirit of the Fermi-liquid theory of weak-coupling superconductors,[8] the Landau parameters being replaced (for our purpose) by the bare interaction V. They comprise the sum of ladder and bubble diagrams, and the Ward identities are fulfilled exactly. This should be contrasted with a recent, related calculation,[9] in which only the first two terms in (8) were considered, leading to incomplete and unphysical results.

The exact solution of (7) and (8) in the strong-coupling, excitonic limit is tedious but straightforward, and details will be given elsewhere.[10] By noting that there is *no* scattering of quasiparticles, but only pair creation and annihilation, we express the diagonal elements of $\delta\hat{n}_{\mathbf{k}}(\mathbf{q})$ in terms of the off-diagonal ones:

$$(\delta\hat{n}_{\mathbf{k}}(\mathbf{q}))_{11}=[\psi_{\mathbf{k}}^{*}(\delta\hat{n}_{\mathbf{k}}(\mathbf{q}))_{21}+\psi_{\mathbf{k}+\mathbf{q}}(\delta\hat{n}_{\mathbf{k}}(\mathbf{q}))_{12}]/[1-n_{\mathbf{k}}-n_{\mathbf{k}+\mathbf{q}}], \tag{9a}$$

$$(\delta\hat{n}_{\mathbf{k}}(\mathbf{q}))_{22}=-[\psi_{\mathbf{k}+\mathbf{q}}^{*}(\delta\hat{n}_{\mathbf{k}}(\mathbf{q}))_{21}+\psi_{\mathbf{k}}(\delta\hat{n}_{\mathbf{k}}(\mathbf{q}))_{12}]/[1-n_{\mathbf{k}}-n_{\mathbf{k}+\mathbf{q}}]. \tag{9b}$$

In the long-wavelength limit $\mathbf{q}\rightarrow 0$, we make the *Ansatz*

$$(\delta\hat{n}_{\mathbf{k}}(\mathbf{q}))_{21}=(\delta\hat{n}_{\mathbf{k}}(\mathbf{q}))_{12}^{*}=\psi_{\mathbf{k}}^{+}e^{-i\Delta\omega t}+(\psi_{\mathbf{k}}^{-})^{*}e^{i\Delta\omega t} \tag{10}$$

and expand $\psi_{\mathbf{k}}^{\pm}$ in terms of the unperturbed exciton wave functions $\phi_{n\mathbf{k}}$, $\psi_{\mathbf{k}}^{+}=\sum_n c_n^{+}\phi_{n\mathbf{k}}$ and $\psi_{\mathbf{k}}^{-}=\sum_n c_n^{-}\phi_{n\mathbf{k}}^{*}$. The coefficients $c_n^{\pm}$ obey then Beliaev's equations[11] in the weak-coupling (of pairs !) limit:

$$(\Delta\omega+i0+\omega_p-E_n^0)c_n^{+}=-\mu E_t\sum_{\mathbf{k}}(1-2n_{\mathbf{k}})\phi_{n\mathbf{k}}^{*}+\sum_m[(\Sigma_{nm}^{\mathrm{H}}+\Sigma_{nm}^{\mathrm{F}}+\Pi_{nm}^{\mathrm{F}})c_m^{+}+(\Sigma_{nm}^{\mathrm{B}}+\Pi_{nm}^{\mathrm{B}})c_m^{-}], \tag{11a}$$

$$(\Delta\omega+i0-\omega_p+E_n^0)c_n^{-}=-\sum_m[(\Sigma_{nm}^{\mathrm{H}}+\Sigma_{nm}^{\mathrm{F}}+\Pi_{nm}^{\mathrm{F}})c_m^{-}+(\Sigma_{nm}^{\mathrm{B}}+\Pi_{nm}^{\mathrm{B}})c_m^{+}], \tag{11b}$$

where

$$\Sigma_{nm}^{\mathrm{F}}=(2\langle n\psi\,|\,I_d\,|\,\psi m\rangle+\langle n\psi\,|\,I_x\,|\,\psi m\rangle)\quad[\Sigma_{nm}^{\mathrm{B}}=(2\langle nm\,|\,I_d\,|\,\psi\psi\rangle+\langle nm\,|\,I_x\,|\,\psi\psi\rangle)]$$

and

$$\Pi_{nm}^{\mathrm{F}}=2\mu E_p\sum_{\mathbf{k}}\phi_{n\mathbf{k}}^{*}\phi_{m\mathbf{k}}\langle\psi\,|\,\mathbf{k}\rangle\quad(\Pi_{nm}^{\mathrm{B}}=2\mu E_p\sum_{\mathbf{k}}\phi_{n\mathbf{k}}^{*}\phi_{m\mathbf{k}}^{*}\langle\mathbf{k}\,|\,\psi\rangle)$$

are exciton Fock (Bogolyubov) self-energies due to *exciton-exciton* (Σ) and *anharmonic exciton-photon* (Π) interactions. Interestingly, the direct exciton interaction I_d appears as a result of screening.

In leading order in the pump intensity (e-h pair density) exciton pair creation and annihilation, as described by the Bogolubov self-energies, can be neglected. The spectrum of collective excitations [eigenenergies of (11)] is then given by

$$\omega_n=[(E_n^{\mathrm{HF}}-\omega_p)^2-|\Sigma_{nn}^{\mathrm{B}}+\Pi_{nn}^{\mathrm{B}}|^2]^{1/2}\simeq E_n^{\mathrm{HF}}-\omega_p, \tag{12}$$

where $E_n^{\mathrm{HF}}=E_n^0+\Sigma_{nn}^{\mathrm{H}}+\Sigma_{nn}^{\mathrm{F}}+\Pi_{nn}^{\mathrm{F}}$ is the renormalized (blue-shifted) exciton energy, as seen by the test beam. The nonlinear susceptibility χ_t takes on the same form as χ_p, except that ω_p is replaced by ω_t+i0 and Σ^{H} by $\Sigma^{\mathrm{H}}+\Sigma^{\mathrm{F}}+\Pi^{\mathrm{F}}$. Again, these results can be easily generalized to account for multiple exciton-exciton scattering. For frequencies ω_t close to or above the lowest exciton resonance, exciton-phonon interactions should ideally be included.

In general, the polarization induced by the test beam is determined by the full $(\delta\hat{n}_{\mathbf{k}}(\mathbf{q}))_{21}$ and consists of two contributions at $\omega_t=\omega_p\pm\omega_n$. Absorption occurs at the renormalized exciton energy $\omega_p+\omega_n$, and gain at $\omega_p-\omega_n$. In the latter process, two pump photons are destroyed and a test photon and renormalized exciton are created, resulting in emission at $\omega_t=2\omega_p-(\omega_p+\omega_n)$. This process is due to the "depletion of the condensate" and thus at least quadratic in the pump intensity.

Most importantly, the coherent excitation produces a contribution Π_{nn}^{F} to the Stark shift and a corresponding bleaching (due to Π_{nm}^{F}, $n\neq m$), which are due to anharmonic exciton-photon interactions. The latter result from the composite nature of excitons, in much the same way as the exciton-exciton interaction. If we consider the (leading) $1s$-exciton contribution to $\Pi_{1s\,1s}^{\mathrm{F}}$, we obtain for the anharmonic contribution to the ac Stark shift

$$\Pi_{1s\,1s}^{\mathrm{F}}\simeq\frac{2|\mu E_p|^2}{E_{1s}^0-\omega_p}\,\frac{|\phi_{1s}(\mathbf{r}=0)|^2}{N_s^{\mathrm{PSF}}}, \tag{13}$$

where N_s^{PSF} is the saturation density due to excitonic phase-space filling. This explains already the recent results obtained on GaAs-$Al_xGa_{1-x}As$ quantum wells[2] quantitatively, without any adjustable parameters. For example, for a pump intensity $\simeq 8\times10^6$ W cm^{-2} and a detuning $E_{1s}^0-\omega_p\simeq 3\times10^{-2}$ eV, Eq. (13) yields a Stark shift $\simeq 1.5\times10^{-4}$ eV, which is in good agreement with the experimental value $\simeq 2\times10^{-4}$ eV. Moreover, it shows once more that excitons behave like N_s^{PSF} two-level atoms, to the extent that exciton-exciton interac-

tions can be neglected. Interestingly, the magnitude of (13) is independent of dimension.

Let us finally note that our theory should also have great impact on various related problems. Its extension to four-fermion pairing is straightforward and shows that the theory of the two-photon generation of biexcitons[5,12] has to be revised. Within the T-matrix approximation, it allows for the *first* first-principles calculations of transport in Bose-condensed atomic systems, such as $H_{\uparrow}$.[13] For Bose-condensed systems, it yields the exact excitation spectrum (the Anderson-Bogolyubov mode) in both the weak- *and* strong-coupling limits, and thus allows for a calculation of the critical temperature for arbitrary coupling strengths.[14]

We wish to thank J. P. Heritage, D. Hulin, A. von Lehmen, S. L. McCall, A. Migus, J. Orenstein, C. M. Varma, and J. E. Zucker for stimulating discussions and collaboration, and G. Mahler for making his work available prior to publication.

[1]A. Mysyrowicz, D. Hulin, A. Antonetti, A. Migus, W. T. Masselink, and H. Morkoc, Phys. Rev. Lett. **56**, 2748 (1986).

[2]A. von Lehmen, D. S. Chemla, J. E. Zucker, and J. P. Heritage, Opt. Lett. (to be published).

[3]D. Fröhlich, A. Nöthe, and K. Reimann, Phys. Rev. Lett. **55**, 1335 (1985).

[4]B. R. Mollow, Phys. Rev. **188**, 1969 (1969), and Phys. Rev. A **5**, 2217 (1972).

[5]H. Haug and S. Schmitt-Rink, Prog. Quant. Electron. **9**, 3 (1984), and J. Opt. Soc. Am. B **2**, 1135 (1985).

[6]See, e.g., V. M. Galitskii, S. P. Goreslavskii, and V. F. Elesin, Zh. Eksp. Teor. Fiz. **57**, 207 (1969) [Sov. Phys. JETP **30**, 117 (1970)]; H. Haug, J. Lumin. **30**, 171 (1985).

[7]P. W. Anderson, Phys. Rev. **112**, 1900 (1958).

[8]O. Betbeder-Matibet and P. Nozières, Ann. Phys. (N.Y.) **51**, 392 (1969).

[9]C. Comte and G. Mahler, to be published.

[10]S. Schmitt-Rink and D. S. Chemla, to be published.

[11]S. T. Beliaev, Zh. Eksp. Teor. Fiz. **34**, 417 (1958) [Sov. Phys. JETP **7**, 289 (1958)], and Zh. Eksp. Teor. Fiz. **34**, 433 (1958) [Sov. Phys. JETP **7**, 299 (1958)].

[12]See, e.g., I. Abram, J. Phys. Soc. Am. B **2**, 1204 (1985); B. Hönerlage, R. Lévy, J. B. Grun, C. Klingshirn, and K. Bohnert, Phys. Rep. **124**, 161 (1985).

[13]A. E. Ruckenstein and L. P. Levy, to be published.

[14]P. Nozières and S. Schmitt-Rink, J. Low. Temp Phys. **59**, 195 (1985).