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Excitonic optical nonlinearity of quantum-well structures in a static electric field

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The excitonic nonlinear optical susceptibility $\chi^{(n)}$, of multiple-quantum-well structures subject to a static electric field normal to the layers is theoretically investigated when the incident photon energy is lower than, but comparable to, the absorption edge. We have taken into account both the effect of giant dipole moment (GDM) and that of virtual charge (VC) of excitons. For $n=2$, the VC effect is absent, and $\chi^{(2)}$ is enhanced solely by the GDM effect, resulting in a value about 10 times larger than that of the bulk. For $n=3$, on the other hand, both the GDM and the VC effects contribute to large $\chi^{(3)}$. The former is dominant near the two-photon resonance, while the latter is dominant near the one-photon resonance. Resulting enhanced values of $\chi^{(3)}$ are 10–10^3 times larger than that of the bulk.

Optical nonlinearities of semiconductors can be greatly enhanced in quantum-well structures (QWS's) over those of the bulk crystals due to various quantum size effects such as confinement of electron wave functions.[1-12] Of particular interest are optical nonlinearities of QWS's in an external static electric field normal to the layers,[9-12] since the wave functions of electron-hole pairs or excitons are greatly deformed, but the overlap integral of electron-hole pairs remains finite.[13] Two different mechanisms of nonlinearities specific to such a system have been suggested. Yamanishi[9] and Chemla, Miller, and Schmitt-Rink[10] independently pointed out that the depolarization field induced by the "virtual charge" (VC) of photoexcited electron-hole pairs gives rise to large optical nonlinearity. On the other hand, the present author[12] pointed out that the "giant dipole moment" (GDM) of excitons also gives rise to large optical nonlinearity. These two mechanisms are *quite different in character*, and they were investigated separately. The VC effect comes from the dipole-dipole interaction between the excitons, whereas the GDM effect is basically independent of any correlations between the excitons.[12]

In this work, we theoretically investigate optical nonlinearities, taking both effects into account. Since excitons are responsible to optical properties of QWS's even at room temperature,[5] and the above two effects become appreciable near the excitonic resonance, we shall consider the excitonic contribution, rather than the contribution from unbound electron-hole pairs, to the nth-order nonlinear optical susceptibility, $\chi^{(n)}$. The incident photon energy is assumed to be lower than the absorption edge, so that *no real exciton population is involved*. This means that the response time is ultrafast *both* for ON and OFF processes,[9-12] in comparison to mechanisms involving real populations.[4,5]

We consider the contribution to $\chi^{(n)}$ from the $1S$ heavy-hole excitons associated with the lowest subbands. The Hamiltonian of the excitons[11] may be represented as

$$H(t)=\sum_{\nu}\varepsilon_{\nu}a_{\nu}^{\dagger}a_{\nu}-F_{dpl}(t)e\sum_{\nu}l_{\nu}a_{\nu}^{\dagger}a_{\nu}-P^{\text{tot}}E(t)\ , \qquad (1)$$

where $a_{\nu}^{\dagger}$ (a_{ν}) denotes the creation (annihilation) operator of the exciton with a quantum number ν, ε_{ν} is the exciton energy in the static bias field, l_{ν} is the GDM,[12] $F_{dpl}(t)$ represents the depolarization field due to the VC,[9-11] $E(t)=E_{\omega}e^{-i\omega t}+\text{c.c.}$ is the electric field of light, and P^{tot} is the total dipole moment operator given by

$$P^{\text{tot}}=e\sum_{\nu}(\mu_{\nu}a_{\nu}^{\dagger}+\text{H.c.})+e\sum_{\nu}l_{\nu}a_{\nu}^{\dagger}a_{n}\ . \qquad (2)$$

Here, μ_{ν} is the transition dipole moment which is, in the same notations as in Ref. 12, given by

$$\mu_{\nu}=-\sqrt{S}\delta_{\mathbf{K}_{\parallel},0}RI_{eh}^{*}\sqrt{2a^{2}/\pi l_{z}}\equiv-\sqrt{S}\delta_{\mathbf{K}_{\parallel},0}M\ .$$

In the above expression, all the vector quantities are assumed to be along the z directions. Thus, calculated results for $\chi^{(n)}$'s given below correspond to their $z,\ldots,z$ tensor components. In actual systems, however, the anisotropy in μ_{ν}, which is inherent to QWS's,[14] can modify the magnitude of tensor components of $\chi^{(n)}$;[12] some components become larger and others become smaller than those for the isotropic case. Therefore, the numerical values of $\chi^{(n)}$ given below should be considered as representative values of various tensor components of $\chi^{(n)}$.

The second term of $H(t)$ gives rise to nonlinearities due to the VC effect.[9-11] This term describes the dipole-dipole interaction between the excitons[12] within the framework of a mean-field approximation. In the last term of $H(t)$, the second term of Eq. (2) multiplied by $E(t)$ represents the nonlinear interaction of the GDM of excitons with the light field. This term, which was disregarded in previous work,[9-11] is the origin of the nonlinear response due to the GDM effect. It should be noted that the rotating wave approximation, used in the previous work,[9-11] would lead to incorrect results for $\chi^{(n)}$'s for the present $H(t)$. For example, the term proportional to $[(E_{ex}-\hbar\omega)(E_{ex}+\hbar\omega)]^{-1}$ appearing in $\chi^{(2)}$ could not be obtained, resulting in disagreement with the correct result given below.

We can diagonalize $H(t)$ by the unitary operator,[11]

$$U(t)\equiv\exp\left[i\sum_{\nu}[x_{\nu}(t)a_{\nu}^{\dagger}+\text{H.c.}\right]\ , \qquad (3)$$

under the self-consistency condition,[9-11]

$$F_{dpl}(t)=-4\pi\langle\phi(t)\,|\,e\sum_{\nu}l_{\nu}a_{\nu}^{\dagger}a_{\nu}\,|\,\phi(t)\rangle/V\ .$$

Here, $|\phi(t)\rangle$ denotes the ground state of $H(t)$, V is the volume of the system, and $x_\nu(t)$ is a c number which satisfies the following nonlinear differential equation:

$$\hbar \frac{d}{dt} x_\nu + i \left[\varepsilon_\nu + 4\pi e^2 l_\nu \sum_{\nu'} l_{\nu'} |x_{\nu'}|^2 - e l_\nu E(t) \right] x_\nu + e\mu_\nu E(t) = 0 \; . \quad (4)$$

The nonlinear terms, i.e., the second and the third terms in the large parentheses come from the VC and the GDM effects, respectively. The polarization induced in the system, $P(t)$, is given by the expectation value of P^{tot}/V for the ground state, $|\phi(t)\rangle = e^{i\theta(t)} U^\dagger(t) |g\rangle$, where $\theta(t)$ is a phase factor and $|g\rangle$ is the ground state for $E(t)=0$. The $P(t)$ is calculated as

$$P(t) = \left[e \sum_\nu (i\mu_\nu x_\nu^* + \text{c.c.}) + e \sum_\nu l_\nu |x_\nu|^2 \right] / V \; . \quad (5)$$

We can easily obtain a solution of Eq. (4) as a power series of E_ω and E_ω^*. Substituting the power series solution of $x_\nu(t)$ into Eq. (5), and retaining resonant terms only, we finally obtain $\chi^{(2)}(2\omega;\omega,\omega)$ and $\chi^{(3)}(\omega;\omega,\omega,-\omega)$ as follows:

$$\chi^{(2)} = C_{\text{GDM}}^{(2)} \times \begin{cases} -1/E_g\Delta, & \hbar\omega = E_{ex} - \Delta, \\ 4/E_g\Delta, & 2\hbar\omega = E_{ex} - \Delta, \end{cases} \quad (6)$$

$$\chi^{(3)} = \begin{cases} -C_{\text{VC}}^{(3)}/\Delta^4 - C_{\text{GDM}}^{(3)}/E_g^2\Delta, & \hbar\omega = E_{ex} - \Delta, \\ 4C_{\text{GDM}}^{(3)}/E_g^2\Delta, & 2\hbar\omega = E_{ex} - \Delta, \end{cases} \quad (7)$$

where

$$C_{\text{GDM}}^{(2)} \equiv e^3 l |M|^2/(L_z + L_B) \; ,$$

$$C_{\text{VC}}^{(3)} \equiv 16\pi e^6 l^2 |M|^4/(L_z + L_B)^2 \; ,$$

$$C_{\text{GDM}}^{(3)} \equiv 4e^4 l^2 |M|^2/(L_z + L_B) \; ,$$

the subscripts "GDM" and "VC" indicate the origins of the corresponding terms, $L_z(L_B)$ is the thickness of a well (barrier) layer, and we have used the relation E_{ex} ($\equiv \varepsilon_\nu$ for $\mathbf{K}_\parallel = 0$) $\approx E_g$ (the band-gap energy of the well) $\gg |\Delta|$ (the detuning energy). The expression of the GDM of the exciton l was given explicitly in Ref. 12.

The above result for $\chi^{(2)}$ agrees with that for $\chi_{zzz}^{(2)}$ obtained using a different method in Ref. 12. It is confirmed that the VC effect is absent in the second-order response. We can also see that no cross terms of the GDM and the VC effects appear up to the third-order response. This can also be easily shown from the nonlinear-response-theoretical expression of $\chi^{(n)}$.

Both $\chi^{(2)}$ and $\chi^{(3)}$ exhibit resonant enhancement both at the one- and the two-photon resonance. We shall focus on $\chi^{(3)}$, since approximately ten times enhancement over the bulk crystal was already demonstrated for $\chi^{(2)}$ in Ref. 12. The calculation of higher-order ($n=4,5,\ldots$) susceptibilities is straightforward, but they will not be discussed here. In Fig. 1, we have plotted $\chi^{(3)}$ for the GaAs/$\text{Al}_x\text{Ga}_{1-x}\text{As}$ multiple QWS, (a) near the one-photon and (b) near the two-photon resonance, for $L_z = 120$ Å, $L_B = 80$ Å, and F_{bias} (the bias field) $= 100$ kV/cm.[12] We shall compare these results with $\chi^{(3)}$ of the bulk GaAs. To the author's knowledge, however, no experimental data

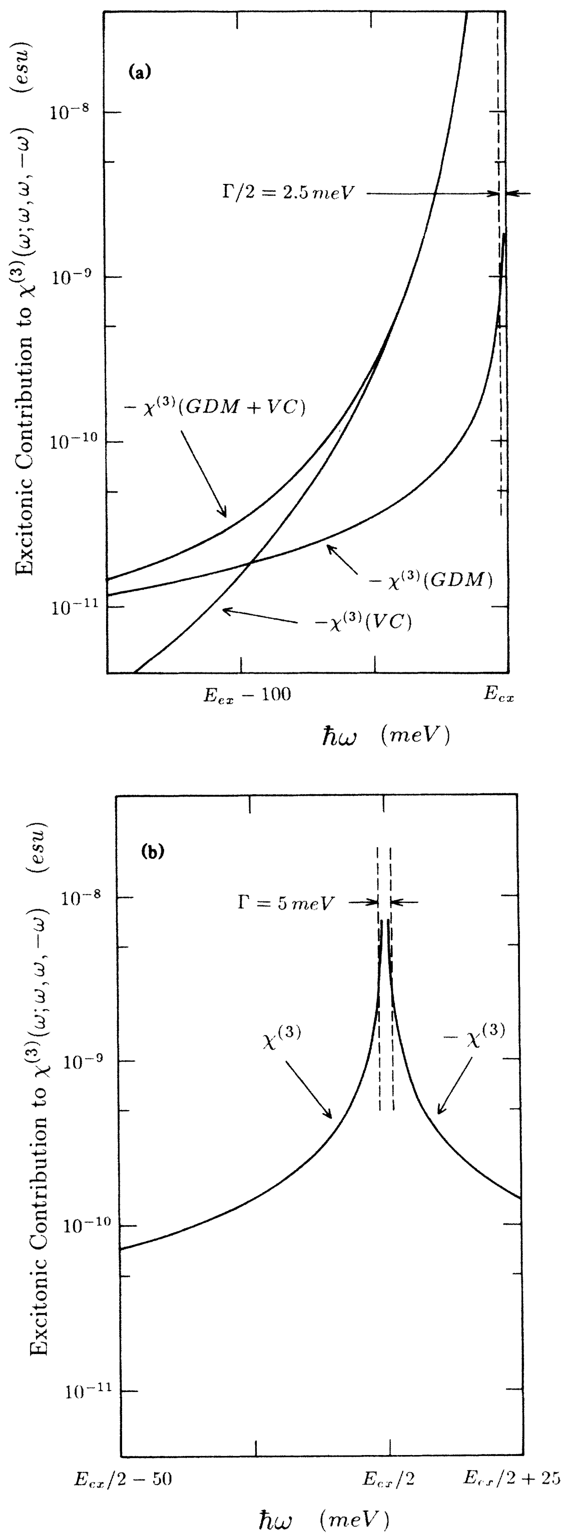

FIG. 1. Excitonic contribution to $\chi^{(3)}(\omega;\omega,\omega,-\omega)$, near the (a) one-photon, and (b) two-photon resonance. In (a), the three solid lines denoted by GDM, VC, and GDM+VC represent the contributions from the giant dipole moment effect, the virtual charge effect, and the sum of both, respectively. In (b), only the GDM term is plotted since the VC term is negligibly small. Note the sign of $\chi^{(3)}$. For example, $\chi^{(3)}$ is antisymmetric about the line $\hbar\omega = E_{ex}/2$ in (b).

of $\chi^{(3)}$ for the above photon energies are available; the existing data are limited to either $\hbar\omega \ll E_{ex}$ (Ref. 15) or $\hbar\omega \gtrsim E_{ex}$.[5] For the latter, nonlinearities due to real exciton population become dominant,[5] so the situation is quite different. For the former, the contribution from free carriers, $\chi_n^{(3)}$, and that from bound or valence electrons, $\chi_b^{(3)}$, are of the same order of magnitude.[15] For higher photon energies as considered here, $\chi_n^{(3)}$ would become small, and the dispersion of $\chi_b^{(3)}$ is expected to be weak except at such very low temperatures that resonant enhancement by *bulk* excitons could become appreciable. Therefore, we shall use $\chi_b^{(3)}$ ($\approx 10^{-11}$ esu) measured at 11.8 μm[15] as $\chi^{(3)}$ of the bulk GaAs.

In the case of one-photon resonance, the dependence of $\chi^{(3)}$ on Δ is different between the GDM and the VC terms, and the VC effect is dominant for $\Delta < 100$ meV, giving rise to $10-10^3$ times enhancement of $\chi^{(3)}$ over the bulk crystal.[9-11] The GDM effect becomes dominant when $\Delta > 100$ MeV, but the magnitude of $\chi^{(3)}$ becomes of the same order, with opposite sign, as that of the bulk. As for the two-photon resonance, on the other hand, only the GDM effect contributes, resulting in $10-10^3$ times enhancement of $\chi^{(3)}$ over the bulk.

It should be noted that the formulas (6) and (7) are valid for $|\Delta| \gtrsim \Gamma/2$ because of our assumption of off resonance,[12] where Γ is the full width of the exciton peak.[5] The dashed lines in Fig. 1 represent the lines, $|\Delta| = \Gamma/2$, where Γ is taken as 5 meV. We shall briefly comment on the case $|\Delta| \lesssim \Gamma/2$. In order to calculate the behavior of $\chi^{(n)}$ for $|\Delta| \lesssim \Gamma/2$, we have to convolute various damping mechanisms. Unfortunately, however, it has not been clarified, even for the *linear* susceptibility, how to convolute them.[13] At present, we can only state that the curves for $|\chi^{(n)}|$, which grow toward the resonance, would drop when $|\Delta| \lesssim \Gamma/2$, and as a result the maximum values of $|\chi^{(n)}|$ would be obtained at $|\Delta| \approx \Gamma/2$. For the one-photon resonance case, however, $\chi^{(n)}$ due to the present mechanism would be masked for $\Delta \lesssim \Gamma/2$ by other enhancement mechanisms[4,5] involving real exciton populations.

In summary, we have theoretically investigated excitonic optical nonlinearities of multiple quantum-well structures in a static electric field normal to the layers, taking into account both the giant dipole moment effect and the virtual charge effect of excitons. The explicit formulas for $\chi^{(2)}$ and $\chi^{(3)}$ are obtained as functions of QW parameters. Numerical results are given for a typical (not optimum) $GaAs/Al_xGa_{1-x}As$ multiple QWS. For $n=2$, the VC effect is absent, and $\chi^{(2)}$ is enhanced solely by the GDM effect, resulting in about 10 times larger value than that of the bulk. This enhancement occurs for $\hbar\omega \approx E_{ex}$ and for $2\hbar\omega \approx E_{ex}$. For $n=3$, on the other hand, both the GDM and the VC effects contribute to large $\chi^{(3)}$. The former is dominant for $2\hbar\omega \approx E_{ex}$, while the latter is dominant for $\hbar\omega \approx E_{ex}$. Resulting enhanced values of $\chi^{(3)}$ are $10-10^3$ times larger than that of the bulk.

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