Field-Induced Optical Nonlinearity Due to Virtual Transitions in Semiconductor Quantum-Well Structures

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A new concept of optical nonlinearity due to virtual transitions in quantum-well structures in electric field, named virtual charge–induced optical nonlinearity, is proposed, and some examples of theoretical results on the nonlinearity given. The switching time of the nonlinearity is expected to be extremely short, 100 fs. The nonlinearity seems to be observable and quite useful for the design of an ultrafast optical gate. A possibility of optical bistability without external optical feedback, based on the proposed nonlinearity, is pointed out.

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Optical nonlinearities of semiconductors are quite interesting aspects, from the standpoint of both fundamental physics and device applications. Particularly, the nonlinearities due to virtual transitions of electrons are very attractive because of their inherent high-speed switching capabilities. For instance, an ultrafast dynamical blue shift (ac Stark effect) accompanied by a strong bleaching in GaAlAs quantum-well (QW) structures has been discovered[1,2] and interpreted in terms of a dressed-exciton model[1,3] in which a coherent and direct-nonlinear interaction of excitons with photons plays an essential role. The aims of this Letter are to propose a new concept of ultrafast optical nonlinearity due to virtual transitions in QW structures in dc electric fields,[4] and to show some examples of theoretical results on the proposed nonlinearity. In the proposed scheme, an asymmetry of the QW structure is essential for the nonlinearity. In this point, the present mechanism is completely different from the previous ones such as the dressed-exciton model.

As is well known, excitonic and subband transition gaps in a QW structure can be shifted to the red by an applied electric field perpendicular to the QW plane. Also, oscillator strengths relevant to the electronic transitions can be decreased or increased by the field, depending on allowed or forbidden transitions at zero field.[5,6] In the QW structure subjected to a dc electric field E_0, perpendicular to the heterojunction plane as shown in Fig. 1, positive and negative electric charges at the subbands (1hh, 2hh, . . . , 1lh, 2lh, . . . in the valence band and $1e$, $2e$, . . . in the conduction band, respectively) are induced by virtual transitions due to an intense pump light with a photon energy $\hbar\omega_p$ far below the fundamental band gap of the QW, $E_{1e\text{-}1hh}$. The induced positive (virtual holes) and negative (virtual electrons) charges may produce a screening field E_s which has an opposite polarity to that of the original dc field. Consequently, one may expect a blue shift of the fundamental gap and changes in the oscillator strengths for a weak signal photon with a photon energy $\hbar\omega_s$. This is the proposed mechanism, named virtual charge–induced optical nonlinearity (VCON), which will be, in more detail, discussed in the following. The response time of the overall processes for a pulsed pump light should be very short, $\simeq 100$ fs, for both the on and off switchings because the electric charges are induced by the virtual processes, and the field cancellation results from the internal charges inside the QW. In other words, the switching characteristic is completely free from lifetime limitation, in contrast with those due to real excitation processes, and also, from free CR–time-constant limitation.

For two collisionless quantum states, designated as 1 and 2, interacting with a classical monochromatic radiation field represented by $E_p\cos(\omega_p t)$, we take the wave function as a linear superposition of $\phi_1(\mathbf{r})u_1(\mathbf{r})$ and $\phi_2(\mathbf{r})u_2(\mathbf{r})$ which are eigenfunctions [$\phi_i(\mathbf{r})$, the envelope function, and $u_i(\mathbf{r})$, the Bloch function] of the unper-

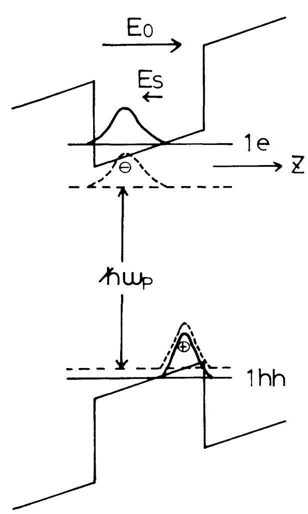

FIG. 1. Quantum-well structure biased by a dc field E_0, and pumped by an off-resonant light, $\hbar\omega_p < E_{1e-1hh}$.

turbed Hamiltonian H_0,[7]

$$\psi(\mathbf{r},t)=a_1(t)\phi_1(\mathbf{r})u_1(\mathbf{r})e^{-i\omega_1 t}+a_2(t)\phi_2(\mathbf{r})u_2(\mathbf{r})e^{-i\omega_2 t}, \tag{1}$$

where $\omega_{1,2}=E_{1,2}/\hbar$. The collision-free assumption can be justified as long as the width of the pump pulse is shorter than the transverse relaxation time (T_2 time) for the off-resonant transition, which should be significantly longer than the T_2 time for a resonant transition.[8] In addition, the virtual population can follow the pump pulse with an intrinsic response time approximately the inverse of the detuning frequency $1/|\Delta_{p,1,2}|$. Under the rotating-wave approximation, the carrier populations represented by $1-|a_1|^2$ (virtual hole), and $|a_2|^2$ (virtual electron) may result in an ensemble average of the dc dipole along the bias-field direction (z axis) induced by the virtual transitions between the levels 1 and 2,

$$\begin{aligned}\langle P_{\mathrm{dc}}\rangle_{1,2}&=[1-|a_1(t)|^2_{\mathrm{DC}}]\langle\phi_1|(+ez)|\phi_1\rangle+|a_2(t)|^2_{\mathrm{dc}}\langle\phi_2|(-ez)|\phi_2\rangle\\&=[2\Omega^2_{p,1,2}/(\omega'_{1,2})^2][\langle\phi_1|ez|\phi_1\rangle+\langle\phi_2|(-ez)|\phi_2\rangle],\end{aligned} \tag{2}$$

where $\omega'^2_{1,2}=\Delta^2_{p,1,2}+4\Omega^2_{p,1,2}$, and $\Omega_{p,1,2}=\langle\phi_1|\phi_2\rangle\langle u_1|er|u_2\rangle_c E_p/2\hbar$ and $\Delta_{p,1,2}=\omega_p-(\omega_2-\omega_1)$ are the Rabi precession and the detuning frequencies, respectively. The subscript dc denotes the dc component of the physical parameters. The angular brackets mean quantum mechanical expectation values. The above-mentioned intuitive consideration that the virtual carriers behave as if they are real ones can be justified by more rigorous treatment.[3,9] In a QW structure, the actual dc polarization can be written as a summation over the contributions from transitions between many quantum states,

$$\begin{aligned}\langle P_{\mathrm{dc}}\rangle_{\mathrm{QW}}=&\sum_{\substack{i=1\mathrm{hh},\,2\mathrm{hh},\,\ldots,\\ 1\mathrm{lh},\,2\mathrm{lh},\,\ldots}}\;\sum_{j=1e,\,2e,\,\ldots}\int(ez)\{|\psi_i(z)|^2-|\psi_j(z)|^2\}dz\\&\times\left[[2\Omega^2_{pij}/(\Delta^2_{pij\mathrm{ex}}+4\alpha\Omega^2_{pij})][2|\phi_{\mathrm{ex}ij}(0)|^2/L_z]+\int_0^\infty[2\Omega^2_{pij}/(\Delta^2_{pij}+4\Omega^2_{pij})](m^*_{rij}/\pi\hbar^2L_z)dE\right],\end{aligned} \tag{3}$$

where

$$\Omega_{pij}=\left\{\int\psi_i^*(z)\psi_j(z)dz\right\}\langle u_i|er|u_j\rangle_c E_p/2\hbar,\quad \Delta_{pij\mathrm{ex}}=\omega_p-\omega_{\mathrm{ex}ij},\quad \Delta_{pij}=\omega_p-(\omega_j-\omega_i)-E/\hbar,$$

$$\alpha=2|\Phi_{\mathrm{ex}ij}(0)|^2/N_s^{\mathrm{PSF}}\sim 4.57,$$

and $\psi_i(z)$, $\psi_j(z)$, $\phi_{\mathrm{ex}ij}(\mathbf{r})$, m^*_{rij}, L_z, N_s^{PSF} are the z-axis envelope wave functions at the subbands i and j, the exciton en-

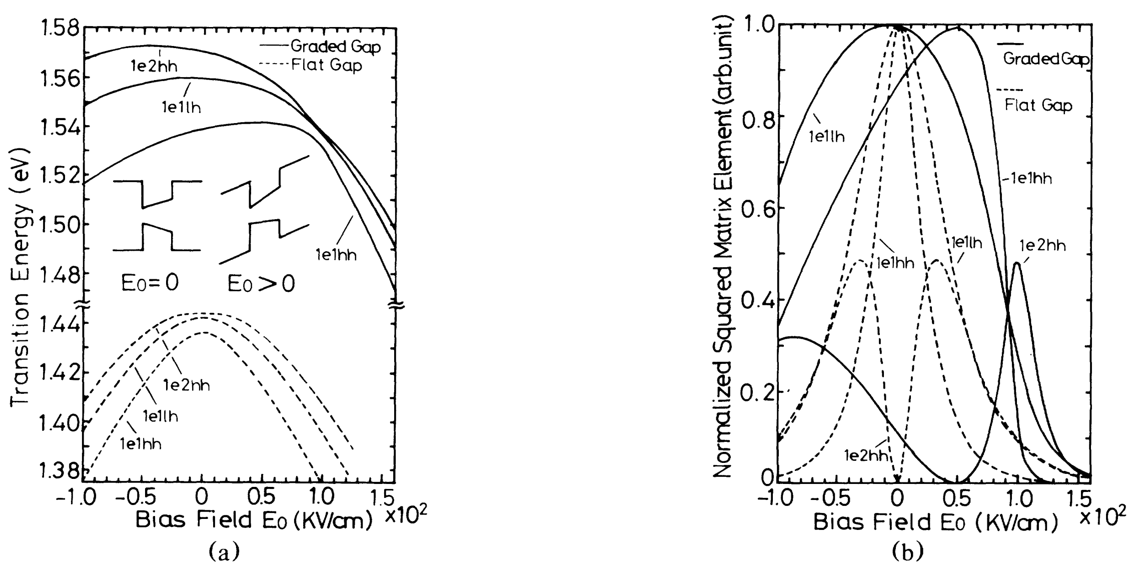

FIG. 2. (a) Transition energies and (b) oscillator strengths as functions of the bias electric field E_0 in a flat-gap GaAs ($L_z=200$ Å)/AlAs (broken lines) and a graded-gap GaAlAs ($L_z=200$ Å)/AlAs (solid lines) quantum-well structure. In the graded-gap quantum-well structure, it is assumed that the Al mole fraction linearly changes with the distance perpendicular to the heterojunction plane, from 0 to 30%.

velope function in the QW plane, the joint-density-of-states mass, the thickness of the QW, and the saturation density of the excitons,[3] respectively. The screening field E_s can be obtained from the relation $E_s = \langle P_{dc} \rangle_{QW}/\epsilon_0 \epsilon_s$, where ϵ_s is the specific dielectric constant in the QW.

If one knows bias-field dependence of the exciton and subband gaps and of the oscillator strengths, as well as the value of E_s, one can easily estimate the amounts of the blue shifts and the variations in the oscillator strengths due to the present mechanism (VCON). We examine two kinds of QW structures: a GaAs/AlAs conventional flat-gap QW (FGQW) with a well-layer thickness of 200 Å, and a $Ga_{1-x}Al_xAs$/AlAs graded-gap QW (GGQW)[10,11] with a well-layer thickness of 200 Å. Figures 2(a) and 2(b) show estimated bias-field dependence of the subband gaps and of oscillator strengths in the QW's. For the GGQW, the bias-field direction was considered to be positive when the conduction-band edge was more tilted, as shown by the inset in Fig. 2(a). The estimated screening field E_s for a pump-power density of 1×10^8 W/cm^2 in the FGQW and GGQW, and for various detuning energies with respect to the fundamental gap $\hbar\Delta_{p0,1e,1hh} = \hbar\omega_p - E_{1e-1hh}$, is shown in Fig. 3. It is worthwhile to note that the E_s is rather insensitive to the detuning energy $\hbar\Delta_{p0,1e,1hh}$ because the contribution of the subband transitions dominates over that of the excitonic transitions for the complete off-resonance condition ($|\hbar\Delta_{p0,1e,1hh}| > 50$ meV). I obtained the amounts of the blue shifts and the variations in the oscillator strengths in both the FGQW and the GGQW. Here, I show the result only for the GGQW because the GGQW showed much more striking features, particularly, the nonlinear changes in the oscillator strengths. Figures 4(a) and 4(b) show the obtained result as well as the exciton blue shifts due to the dressed-exciton mechanism,

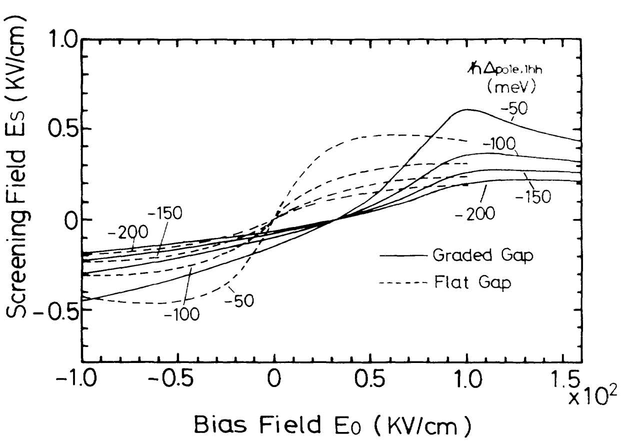

FIG. 3. Screening field E_s due to virtual transitions as functions of the bias field E_0 for a power density of pump light of $I_p = 1\times10^8$ W/cm^2 with various detuning energies, $\hbar\omega_{p0,1e,1hh} = -50$ to -200 meV, in the flat-gap (broken lines) and the graded-gap (solid lines) quantum-well structures.

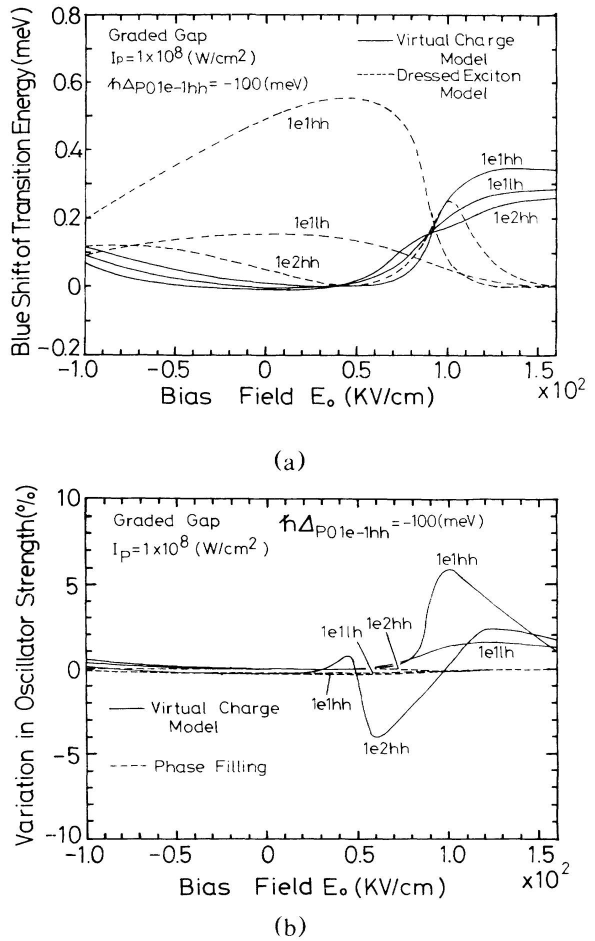

FIG. 4. (a) Blue shift of transition energies and (b) nonlinear variations in oscillator strengths due to the screening field E_s, as functions of the bias field E_0 for a pump-power density of 1×10^8 W/cm^2 and for a detuning energy of -100 meV in the graded-gap quantum well. The variations in oscillator strengths are percentage changes for the oscillator strengths, shown in Fig. 2(b), at each bias field E_0. The blue shifts due to dressed-exciton model, for the pump light with polarization parallel to the QW plane, and the bleachings in oscillator strengths, due to phase-space filling caused by the virtual transitions, are also shown in (a) and (b), respectively.

estimated with Eq. (13) of Ref. 3, and the variations (negative value, the bleaching) in the oscillator strengths due to virtual phase-space filling, represented by $2|a_2(t)|_{dc} = 4\Omega_{pij}^2/(\Delta_{p0ij}^2 + 4\Omega_{pij}^2)$. One can expect significant blue shifts due to the present mechanism (VCON), comparable to those due to the dressed-exciton one. The nonlinear changes in the oscillator strengths sensitively depend on the bias field E_0, taking a maximum value, 5.8% (1e-1hh transition), at a bias field of 100 kV/cm for a pump-power density of 1×10^8 W/cm^2 and a detuning energy $\hbar\Delta_{p0,1e,1hh}$ of -100 meV, and dominating over the bleaching due to the virtual phase-space filling at the same field.

The pump-power–induced increases in the oscillator strengths, in principle, indicate a possibility of optical bistability with no mirrors, cavities, or other external optical feedback, which is similar to optical bistability from increasing absorption.[12] The feedback mechanism might be expected to be as follows: As the pump power exciting the virtual carriers increases, the oscillator strengths relevant to the lowest subband and excitonic transitions may increase because of the field screening due to the virtual charges. The increasing oscillator strengths may result in more excitation of virtual charges, which in turn makes even more increase in the oscillator strengths and so on. Thus, a positive feedback is established. This is particularly true for a large detuning energy because, in such cases, the pump-power–induced blue shift is still smaller than the detuning energy. For quantitative discussions on this class of bistability, we have to consider surface charge density consisting of the polarized virtual charges and dielectric polarization; $Q=\epsilon_0\epsilon_s E_s+\epsilon_0\epsilon_s E_0$. If the surface charge as a function of the bias field E_0 has a negative slope, one may expect to have bistable points for a fixed amount of the charge. In order to realize the feedback mechanism with a reasonable pump-power density, $\simeq 10^9$ W/cm^2, a larger E_s as well as a smaller blue shift are required. It is quite interesting to explore QW structures, more suitable for such an aim.

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Note added.— After the submission of this paper, the same idea was discussed by Chemla, Miller, and Schmitt-Rink at the 1987 International Quantum Electronics Conference at Baltimore. See the following Letter.[13]

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