Optical Quantum-Confined Stark Effect in GaAs Quantum Wells

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The dynamical coupling of the first two conduction-band sublevels in an 84.5-Å GaAs multiple-quantum-well structure by an intense CO_2 laser pulse leads to drastic changes in the absorption spectrum of the lowest excitons. In order to observe this new effect, the polarization of the pump laser has to be perpendicular to the plane of the layers. From the analysis of the experimental results in terms of a three-level nonlinear susceptibility the energy difference of the first two conduction-band sublevels and their transition dipole moment are determined to be 110.3 meV and 14 $e\cdot$Å, respectively.

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Nonlinear optical properties of multiple quantum wells (MQW's) are of great interest not only because of new physical properties but also because of potential applications in electro-optical devices. The electric-field dependence of the optical absorption near the band edge is extensively studied. For reviews, we refer to publications by Miller and co-workers.[1] There is a distinct difference of the field dependence of exciton absorption for fields parallel and perpendicular to the quantum-well layers. In both cases one observed large changes in the absorption near the exciton peaks. In the parallel-field case, the excitons broaden and disappear at fields of about 10^4 V/cm; in the perpendicular configuration, however, the excitons shift to lower energies by as much as 2.5 times their zero-field binding energy, remaining resolved up to 10^5 V/cm. This unique effect is due to the confinement of the carriers which inhibits the exciton field ionization. This effect is called the quantum-confined Stark effect (QCSE).[2] Besides this dc Stark effect, the ac or optical Stark effect in semiconductors has gained increasing interest recently. In ac Stark experiments, one has an additional degree of freedom as compared with the dc case. Besides the intensity which is connected to the electric-field strength, one can also tune the photon energy of the pump field which can lead to a drastic resonance enhancement.

The first observation of a resonant optical Stark effect in a bulk semiconductor was reported by Fröhlich, Nöthe, and Reimann.[3] The authors observed a dynamical coupling of the $1S$ and $2P$ exciton with the use of a high-power tunable CO_2 laser. This excited-state optical Stark effect has to be contrasted with the dynamical coupling of the ground state to, e.g., the first exciton, which was recently demonstrated for MQW's by Mysyrowicz *et al.*[4] and by Von Lehmen *et al.*[5] Both groups observed an instantaneous blue shift of the $n=1$ heavy-hole (HH) and light-hole (LH) exciton resonances induced by a high-power pump beam tuned to as much as 40 meV below the first resonance. The authors interpret this result as an optical Stark effect due to a virtual pumping from the ground state to the lowest exciton states (HH and LH). Schmitt-Rink and Chemla[6] present a theory of this effect, taking into account the "internal structure" of the excitons as "composite particles made from electrons and holes." Because of the expected resonance enhancement, Mysyrowicz *et al.*[4] and Von Lehmen *et al.*[5] interpret the observed effect as a two-level interaction, which is assumed to be a good approximation in the small-excitation regime.[5] For a two-level interaction [Γ_1 ground state and $\Gamma_5(x,y)$ excited states in D_{2d}], one should observe a pronounced dependence of the Stark effect on the relative polarization of the pump and probe laser. As one is dealing with a virtual excitation of one of the Γ_5 components by the pump laser, one expects an effect only if the same component is probed and no effect if the other (orthogonal) component is probed. Von Lehmen *et al.*[5] do not mention a polarization dependence, whereas Mysyrowicz *et al.*[4] explicitly state that they have not observed a dependence on the relative polarization (in plane polarizations). This inconsistency is certainly of great interest, because it might point to the existence of a very fast relaxation process between the orthogonal exciton components [$\Gamma_5(x)$ and $\Gamma_5(y)$].

In addition to the in-plane experiments it is certainly an interesting question to ask whether experiments with pump polarization *perpendicular* to the quantum wells lead to qualitatively different results comparable to the dc QCSE. In this Letter we report first results of an optical QCSE. As a result of the confinement of electrons and holes in the conduction- and valence-band potentials, respectively, one gets the well-known sublevel structure. The dynamical coupling of the $n=1$ and $n=2$ electron sublevels by an intense laser field leads to drastic field-induced effects. As in the dc QCSE the polarization of

the pump field has to be perpendicular to the layers, since the giant dipole matrix element between the $n=1$ and $n=2$ levels vanishes for in-plane polarization. As pointed out, the optical QCSE provides the unique possibility of resonance tuning. For our experiments we have chosen a multiple-quantum-well structure with GaAs layers of 84.5 Å (60 wells with barriers of 230-Å AlGaAs). For this width the energy difference between the first two sublevels in the conduction band is expected to be close to the photon energy of the CO_2 laser ($\hbar\omega = 0.117$ eV).

The experimental setup is shown schematically in Fig. 1. The central part is an active-mode-locked yttrium-aluminum-garnet (YAlG)-dye laser system, which is described in detail by Frohlich, Riemann, and Wille.[7] The laser provides 60-ps dye pulses of 0.1 mJ and 150-ps YAlG pulses (1.06 μm) of 2 mJ. The tunable dye pulses of 0.3-meV spectral width are attenuated by about 10^8 to be used as a probe laser. The 1.06-μm pulses are further amplified to 10 mJ and then used for plasma switching of a transversely excited atmospheric CO_2 laser, as was first proposed by Alcock, Corkum, and James.[8] The 150-ns pulse of the high-power CO_2 laser (0.4 J per pulse) is incident at Brewster's angle with perfect horizontal polarization on a germanium plate. With the use

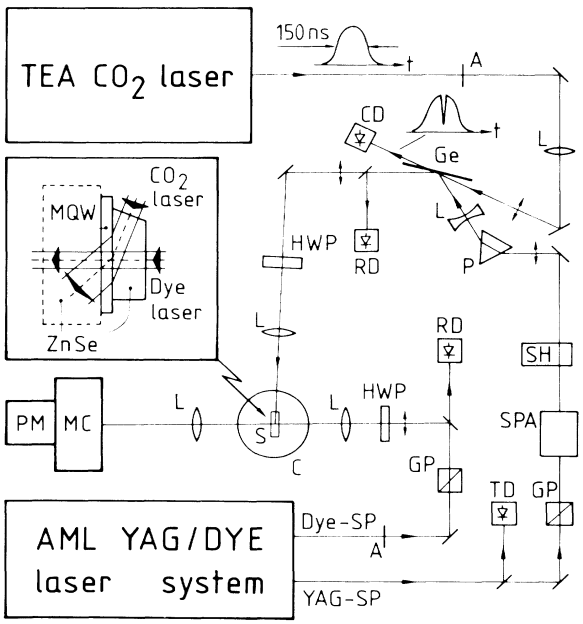

FIG. 1. Block diagram of experimental setup. AML YAG/DYE, active mode-locked YAlG-pumped dye laser system; TEA CO_2, transversely excited atmospheric CO_2 laser; SP, single pulse; GP, Glan polarizer; SPA, single pulse amplifier; SH, shutter; HWP, half-wave plate; P, prism; C, cryostat; S, sample; MC, monochromator; PM, photomultiplier; L, lens; A, attenuator; Ge, germanium plate; TD, trigger diode; RD, reference diode; CD, control diode. Inset: Schematics of optical coupling of CO_2 and dye laser pulse into the MQW.

of a special electronic synchronization system we succeeded in adjusting the YAlG pulse to the maximum of the rather long CO_2 laser pulse with a jitter of less than 15 ns. The YAlG pulse excites a plasma in the germanium plate, which leads to an instantaneous rise of the reflectivity for the 10.6-μm pulse. The finite lifetime of the plasma led to a pulse width of about 250 ps for the 10.6-μm pulse as was measured directly by time-resolved sum-frequency generation in a ZnSe crystal. The shortening of the high-power CO_2 pulse was necessary in order to avoid thermal effects which lead to a red shift of the resonances. The suppression of the long pulse by a careful Brewster alignment was better than 1 in 500. The polarization direction of both lasers can be set by the use of half-wave plates.

As pointed out, it is of special interest to have a large component of the pump beam (10.6-μm pulse) with polarization perpendicular to the layers of the MQW. We used a MQW of about 3×4 mm^2, where the GaAs substrate was totally removed by selective etching. The MQW is mounted on a ZnSe plate. As shown in Fig. 1, we coupled the 10.6-μm pulse through a ZnSe prism into the MQW. The prism is pressed onto the MQW to achieve optical contact. With the use of the ZnSe prism, the intensity of the component with polarization perpendicular to the layers is enhanced by a factor of 5 as compared to a direct coupling into the MQW. This can be easily seen by applying Fresnel's formulas [n(ZnSe) $=2.4$, n(MQW) $=3.3$, $\alpha=70°$].

Experimental results are presented in the upper part of Fig. 2. For both polarization directions of the CO_2 laser, we have measured the transmission of the MQW for the low-power dye laser ($T_\parallel$ and $T_\perp$). For comparison, the unperturbed transmission T_0 is also shown. Comparing $T_\parallel$ and $T_\perp$ one recognizes a pronounced polarization dependence. For $T_\parallel$, one gets a small blue shift which can be compared to similar results of Mysyrowicz *et al.*[4] and Von Lehmen *et al.*[5] For $T_\perp$, however, one gets a qualitatively different effect. Besides a blue shift, one observes an induced absorption on the low-energy side (around 1.55 eV) which is not seen in the $T_\parallel$ configuration. Because of our experimental conditions (refraction angle in MQW approximately 43°), the $T_\perp$ spectrum contains contributions from both polarizations (parallel and perpendicular to the MQW layers), whereas $T_\parallel$ contains only information on the polarization component parallel to the layers.

This pronounced polarization dependence suggests that there is a qualitative difference in the nonlinear couplings for pump polarization parallel and perpendicular to the layers. The blue shift *and* the new induced absorption on the low-energy side of the perpendicular spectrum can be quantitatively described by a nonlinear susceptibility of a three-level system, where two of these levels are dynamically coupled by a strong laser field. In our case we expect a strong coupling between the $n=1$ and $n=2$ sublevels in the conduction band whose energy

separation is close to the photon energy of the CO_2 laser (0.117 eV) for the 84.5-Å MQW. The resonance enhancement for the 0.117-eV CO_2 line was proved by tuning the laser to another strong line at 0.130 eV. In this case the characteristic spectrum of the perpendicular polarization was not seen anymore.

Recently Saikan *et al.*[9] have investigated the variation of the inverse Raman spectrum near resonance. They derive expressions for the overall susceptibility of a three-level system if two of these levels are dynamically coupled by a strong laser field. The absorption constant for the tunable dye laser (photon energy E) in the presence of the high-power CO_2 laser (photon energy $\hbar\omega$) is proportional to the imaginary part of the overall susceptibility. Considering two oscillators (HH and LH transitions), we get the following expression for the overall susceptibility:

$$\chi^{NL}(E,V_{12})=\sum_{\nu=\mathrm{H,L}}\frac{N|\mu_{0\nu\mathrm{H}}|^2}{\epsilon_0}\frac{E_{2\nu}-(E+\hbar\omega)-i\Gamma_{2\nu}}{(E_{\nu\mathrm{H}}-E-i\Gamma_{\nu\mathrm{H}})[E_{2\nu}-(E+\hbar\omega)-i\Gamma_{2\nu}]-|V_{12}|^2}, \tag{1}$$

where the two terms (ν=L,H) refer to the heavy and light holes, N is the density of oscillators, and $\mu_{0\mathrm{HH}}$ and $\mu_{0\mathrm{LH}}$ are the dipole matrix elements for the transition from the ground state to the HH exciton and LH exciton, respectively. E_{HH}, E_{LH} and $E_{2\mathrm{H}}$, $E_{2\mathrm{L}}$ are the values for the transition energies from the valence band ($n-1$, HH and LH levels) to the $n=1$ and $n=2$ conduction-band sublevels, respectively. Γ_{HH}, Γ_{LH}, $\Gamma_{2\mathrm{H}}$, $\Gamma_{2\mathrm{L}}$ represent the damping of the excitons. V_{12} is the transition dipole moment between the $n=1$ and $n=2$ sublevels multiplied by the electric-field strength of the CO_2 laser. The resonance coupling is the same for both oscillators, since the high-power laser is close to resonance with the first two sublevels in the conduction band, whereas the HH and LH excitons are distinguished by their valence-band sublevels only.

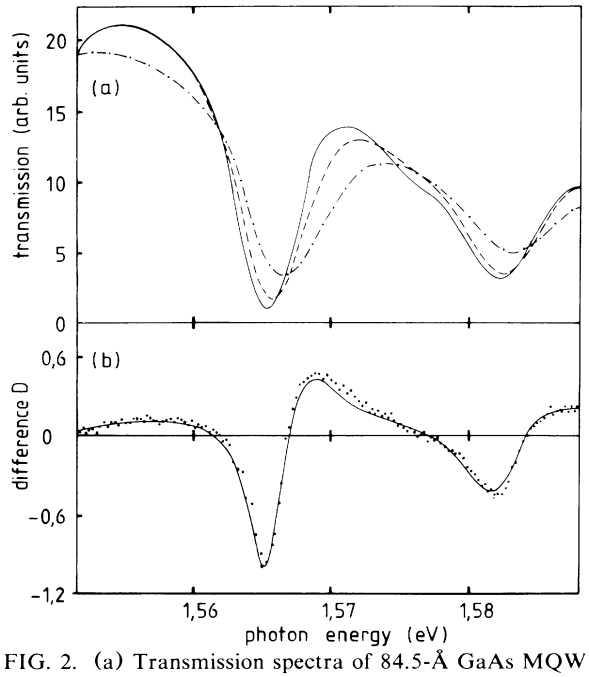

FIG. 2. (a) Transmission spectra of 84.5-Å GaAs MQW at 20 K under different conditions of CO_2 laser irradiation: solid line, transmission T_0 without CO_2 pulse; dashed line, transmission $T_\parallel$ with polarization of CO_2 pulse parallel to the layers; dash-dotted line, transmission $T_\perp$ with polarization of CO_2 pulse parallel and perpendicular to layers. (b) Difference spectrum D calculated from the experimental results (filled circles) and fit by Eq. (2) (full line).

In the lower part of Fig. 2 we show the change in absorption induced by the CO_2 laser which is proportional to the imaginary part of

$$\Delta\chi(E,V_{12})=\chi(E,V_{12})-\chi(E,0). \tag{2}$$

The experimental points are gained from the spectra shown in the upper part of Fig. 2. As pointed out, we get in the perpendicular configuration polarization components of the CO_2 laser parallel and perpendicular to the layers. In our three-level model, as described by Eq. (1), we consider only the perpendicular component. The difference spectrum in the lower part (experimental points) is corrected to show only the nonlinear coupling of the perpendicular component. For this correction the relative intensities for the parallel component in the two polarization configurations have to be considered. If one assumes that the nonlinear effect for the parallel component is proportional to the intensity of the CO_2 laser ($\chi^{(3)}$ effect), one gets for the difference spectrum

$$\Delta\chi\simeq D=\ln(T_0/T_\perp)-(0.51/0.76)\ln(T_0/T_\parallel).$$

The correction factor (0.51/0.76) is calculated with the use of Fresnel's formulas. The solid line in the lower part of Fig. 2 is a fit to the experimental points by Eq. (2). From the fit we derive

$$\Delta E=\hbar\omega-(E_{2\mathrm{H}}-E_{\mathrm{HH}})=\hbar\omega-(E_{2\mathrm{L}}-E_{\mathrm{LH}})$$
$$=6.6\ \mathrm{meV},$$

and for the matrix element $|V_{12}|=5.9$ meV. The damping constants Γ_{HH} and Γ_{LH} are taken from the transmission spectrum T_0 (upper part of Fig. 2). The damping constants $\Gamma_{2\mathrm{H}}$ and $\Gamma_{2\mathrm{L}}$ are set equal to 7 meV, which is about $4\Gamma_{\mathrm{HH}}$. There is one further scaling factor which takes into account the relative oscillator strength of the HH and LH excitons. From $\Delta E=6.6$ meV and $\hbar\omega=116.9$ meV, one gets for the energy separation between the $n=1$ and $n=2$ sublevels $E_{2\mathrm{H}}-E_{\mathrm{HH}}=E_{2\mathrm{L}}-E_{\mathrm{LH}}=110.3$ meV. The matrix element V_{12} can be calculated if the dipole matrix element $e\langle z\rangle$ and the

electric-field strength of the CO_2 laser are known. From the experimentally determined intensity of the CO_2 laser, we derive for the electric-field strength of the component perpendicular to the layers (z component) $E_\perp = (4.2 \pm 0.5) \times 10^4$ V/cm. For an infinite well depth, the dipole matrix element $e\langle z \rangle$ for the $n=1$ and $n=2$ transition is given by $e\langle z \rangle = eL_z \times 16/9\pi^2$ (West and Eglash[10]). For a well width $L_z = 84.5$ Å, we get $\langle z \rangle = 15.2$ Å. With the use of $E_\perp$, we calculate $V_{12} = 6.4$ meV, which is close to our fit parameter $V_{12} = 5.9$ meV. The agreement is quite good if one considers the uncertainty of $E_\perp$. The systematic deviations of the theoretical curve from the experimental points around 1.57 eV might be due to higher excitons and the continuum of the HH transition, which are not considered in the two-oscillator model.

We have looked for a dependence of the dynamical Stark effect on the polarization of the probe laser, which can be changed only in the plane of the layers. The $T_\perp$ signal (see Fig. 2) does not depend on the polarization of the dye laser as expected from symmetry considerations. For the $T_\parallel$ signal, we also got no polarization dependence. This fact cannot be deduced from mere symmetry arguments as discussed in the introduction. We conclude that the blue shift in the $T_\parallel$ spectrum is caused in our case by a nonlinear interaction of the lowest exciton states with higher states. Since higher states (continuum states) of different symmetry are certainly available, no pronounced polarization dependence is expected.

In conclusion we have demonstrated for the first time the optical QCSE, which is due to a resonant coupling of the lowest conduction-band sublevels by a high-power laser. The nonlinear spectrum shows a new induced absorption on the low-energy side of the exciton resonances, which is clearly seen below the HH exciton. In our case, one expects an increase of the resonance effect ($\Delta E \rightarrow 0$) for a slightly smaller well width. Further measurements with other structures are planned to study these resonance effects. Since the optical Stark effect causes an instantaneous change in transmission one can think of possible applications. As seen in Fig. 2, one gets a drastic modulation of a laser beam of about 1.565 eV, which is induced by a laser beam of much smaller photon energy (0.117 eV).

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