Observation of optical Stark effect in InGaAs/InP multiple quantum wells

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We report an experimental observation of a blue shift in the $n = 1$ heavy-hole exciton line of $In_{0.53}Ga_{0.47}As/InP$ multiple quantum wells resulting from a picosecond photoexcitation in the transparent spectral region. The temporal response of this shift follows the excitation and it is attributed to the optical Stark effect. The shift was measured to be 0.19 meV for an incident light with a photon energy 20 meV below the exciton peak and with a 10-MW/cm^2 intensity.

Recently, the observation of an optical Stark effect in bulk and multiple quantum well (MQW) GaAs induced by laser radiation at a photon energy below the fundamental absorption edge was reported.[1,2] This effect manifests itself by a fast (< 1 ps) blue shift in the $n = 1$ exciton lines due to the electric field amplitude associated with the optical wave. Since it does not rely on the generation of real carriers, the temporal response of this "virtual" process essentially follows the laser temporal profile for both rise and recovery and depends solely on the instantaneous intensity rather than on the integrated energy of the excitation. This behavior is different from that resulting from the excitation of real carriers, which depends on the pulse energy and shows recovery time of several nanoseconds. Here, we report the first observation of the optical Stark effect in $In_{0.53}Ga_{0.47}As/InP$ MQW structures, where the InGaAs active layers have an alloy structure and fundamental absorption edges in the 1.5 μm wavelength region.

We used two InGaAs/InP MQW samples grown by chemical beam epitaxy.[3] The samples had undoped thin layers of InGaAs (well) alternating with InP (barrier) on InP substrates.[4] The well thickness, the barrier thickness, and the number of wells were 80 Å, 200 Å, and 50, respectively, for the first sample, and 100 Å, 150 Å, and 100 for the second. Figure 1 shows the absorption spectra of the first sample at 15 and 300 K. The linewidth γ(FWHM) of the $n = 1$ heavy-hole exciton peak is 8.3 and 15.8 meV at the two temperatures, respectively.[5] Its temperature dependence can be fitted in an optical phonon broadening model by the following expression: $\gamma = \gamma_0 + \gamma_1/[\exp(E_{ph}/kT) - 1]$, where $E_{ph} = 32$ meV is the optical phonon energy, $\gamma_0 = 8.3$ meV, and $\gamma_1 = 20$ meV. The low-temperature linewidth γ_0 is approximately triple that of a single quantum well obtained from the photoluminescence measurement.[6] This indicates the samples may have some concentration and well-thickness fluctuations.

In the experiments a nondegenerate pump-and-probe scheme using two tunable picosecond color center lasers (Tl:KCl) was used. The lasers were synchronously pumped by a 1.06-μm mode-locked Nd:YAG laser with a 100-MHz pulse repetition rate. The pulse duration and the tuning range of the lasers were 10 to 15 ps (FWHM) and between 1.45 and 1.57 μm, respectively. The frequency of the pump laser was tuned far below the $n = 1$ heavy-hole exciton peak. The spectral change of this peak due to the off-resonant pump was monitored by tuning the probe wavelength in its vicinity and measuring $\Delta T/T$ (T being the probe transmission and ΔT being the change in T due to the presence of the pump). In the perturbation and thin-sample limits, $\Delta T/T$ is equal to $-\Delta\alpha l$, where $\Delta\alpha$ is the change in absorption coefficient (α) and l is the thickness of the InGaAs. An InGaAs detector in conjuction with two choppers and two lock-in amplifiers was used in the detection.[7]

The pump and probe pulse energies were 0.5 nJ and < 1 pJ, respectively. The two beams were incident at an angle with respect to each other such that they were spatially separated after passing the sample. A monochromator was placed in front of the detector to further suppress the scattered light from the intense pump. The samples were cooled to 80 K to reduce excitation of free carriers by the phonon-assisted mechanism. Finally, two Faraday isolators made of YIG crystals were placed between the two color center lasers and the rest of the experimental setup to avoid possible artifacts due to feedback.

A change in absorption $\Delta\alpha$ at the exciton resonance can be induced by the saturation and broadening of the exciton line and by the optical Stark effect. The first two mechanisms are caused by the real carriers and therefore last longer than several nanoseconds. Since the optical Stark effect is instan-
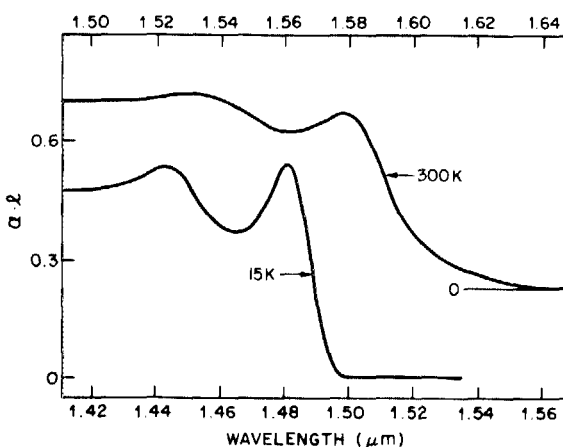

FIG. 1. Absorption spectra of the InGaAs/InP MQW sample at 15 and 300 K.

taneous, it can be distinguished from the carrier effect by measuring the time-resolved $\Delta\alpha$ by scanning the pump-probe delay time. In addition, the spectral response of $\Delta\alpha$ due to the optical Stark effect in the case of an off-resonant pump should follow the derivative of the absorption spectrum. Whereas $\Delta\alpha$ spectrum due to the carriers has a shape similar to the exciton absorption spectrum in the low-density limit.

Figure 2 shows a typical series of delay-time scans of $\Delta T/T$ for the first sample with the probe wavelength as a parameter. Similar results were obtained for the second sample. A positive (negative) sign was designated for the delay time when the pump arrived earlier (later) than the probe. The pump wavelength was detuned by 20 meV below the exciton line. At the exciton peak [Fig. 1(a)], a stepwise curve is shown with the rise occurring at zero delay. The rise time is about the correlation width of the two pulses. This curve is essentially the same as that found in a previous experiment,[7] where the pump was also on resonance with the exciton peak (i.e., the pump and the probe were degenerate) and hence was strongly absorbed. In both cases, ΔT was induced by the saturation and broadening of the exciton line through photogenerated carriers. The only difference is in the direct absorption versus the bandtail absorption for generating the carriers.[8] The nonzero value of ΔT at the negative delay is due to the carrier accumulation: The carrier lifetime is 11 ns for this sample and the carriers generated by one pulse do not decay completely by the arrival of the next pulse 10 ns later. The electron-hole pair density generated by a single pump pulse was deduced[7,8] to be 5.7×10^{15} cm^{-3}, which has been corrected for the accumulation factor of 1.7. The corresponding absorption coefficient was 8.8 cm^{-1}.[8]

The probe pulse, when on resonance with the exciton, created about 10^{16} cm^{-3} carriers.

At longer probe wavelengths, Figs. 2(b)–2(f) show a fast component emerging at the zero delay and superimposed on the stepwise curve. This fast peak, which follows the pump pulse temporal structure and shows an increase in the probe transmission, is attributed to the optical Stark effect. One notices that the long-lived component of $\Delta T/T$ changes sign (i.e., induced absorption) in Figs. 2 (c)–2(g). This is due to the broadening effect, which overcomes the saturation effect at the wing of the exciton line and increases the absorption.

In the following, we determine the amount of $\Delta T/T$ due to the optical Stark effect in Fig. 2 by subtracting the contribution due to the carrier effect. The latter was deduced by measuring $\Delta T/T$ under the conditions that the pump wavelength was tuned inside the absorption region and the pump intensity was reduced by orders of magnitude such that the same density of carriers as in the off-resonant pump case was excited. As a result, the fast peaks in Fig. 2 disappeared and the stepwise $\Delta T/T$ curves were seen at all probe wavelengths. An example is shown by the dashed curve in Fig. 2(c), in which the arrows indicate the difference between the solid and dashed curves at zero delay. Thus, the $\Delta T/T$ spectrum due to the optical Stark effect alone is obtained and is shown in Fig. 3. The low-intensity absorption spectrum and its derivative are also shown. It is evident that the data points in Fig. 3 follow the derivative of the absorption line shape reasonably well. This depicts that the fast component resulted from the shift of the line. The shift $\hbar\delta\omega$ was deduced to be 0.19 meV from the expression $\Delta\alpha = d\alpha/d\omega\times\delta\omega$.[9]

In the short-wavelength (high-energy) portion of a heavy-hole exciton, one would expect an instantaneous reduction (i.e., a fast dip) in $\Delta T/T$ at zero delay. However, no dip could be resolved and no long-lived induced absorption was seen. We note that the high-energy portion of the heavy-hole exciton is overlapped with its energy band gap and the light-hole exciton. The absorption spectrum in that region is rather flat (see Fig. 3). Presumably, the dip due to the heavy-hole exciton could be canceled by the peak from the light-hole exciton. Therefore, the dominant long-lived signal could easily block the weak fast signal if there is any.
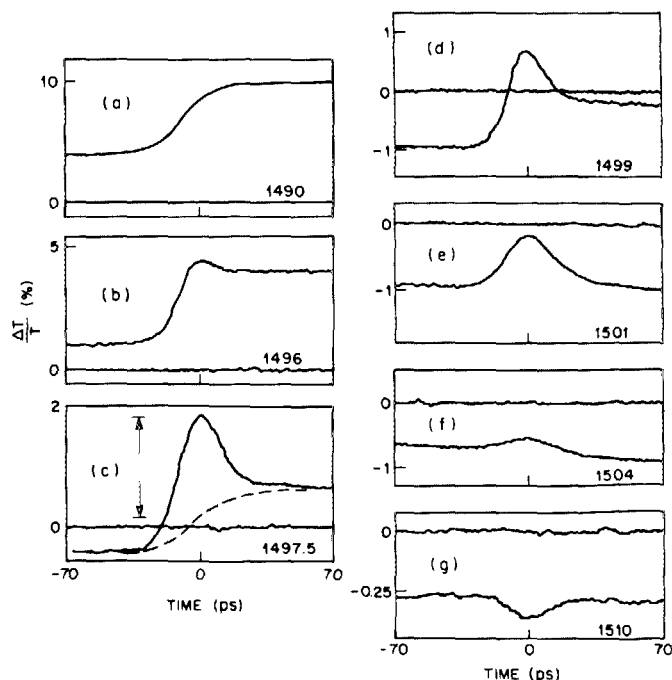

FIG. 2. Series of delay-time scans of $\Delta T/T$ using the probe wavelength as a parameter (expressed in nanometers in the figures). The zero base lines were obtained by blocking the pump beam. The pump wavelength was 1527 nm. The arrows in (c) indicate the difference between the solid and the dashed curves at zero delay.
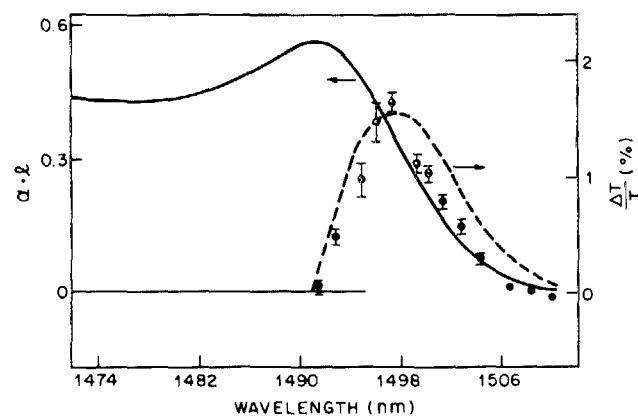

FIG. 3. Solid curve is the absorption spectrum at 80 K. Dashed curve is the calculated derivative of the solid curve. The data points are the $\Delta T/T$ signal due to the instantaneous shift at zero delay.

Schmitt-Rink and Chemla[10] predicted the shift $\hbar\delta\omega$ of the exciton line as following:

$$\hbar\delta\omega \sim 2\frac{|\mu E|^2}{\hbar\Omega}\frac{|U(r=0)|^2}{N_s}, \tag{1}$$

where μ is the interband dipole moment, E is the electric field of the pump beam, Ω is the detuning between the pump and the exciton peak, N_s is the saturation density due to the phase space filling, and $U(r)$ is the exciton wave function $[|U(r=0)|^2 = 1/2\pi a_{2D}^2$, a_{2D} being the two-dimensional exciton Bohr radius]. $|\mu|^2$ can be approximated to be $e^2\hbar^2/4m^*E_g$ assuming a simple two-band model, where m^* is the reduced effective mass of the electron-hole pair, e is the electron charge, $\hbar$ is Planck's constant, and E_g is the energy gap. The first part on the right-hand side of Eq. (1) is exactly the shift found for an atomic two-level system and the second part is of the order unity and can be regarded as the renormalization factor of this atomic shift for the ground-state excitonic transition. Using the known experimental parameters the shift $\hbar\delta\omega$ was calculated to be 0.74 meV for $I = 10$ MW/cm^2 and $\hbar\Omega = 20$ meV. The agreement is satisfactory considering that there exist more than 10^{16} cm^{-3} excess electron-hole pairs in the experiments, which would have partially saturated and broadened the exciton line. In addition, the transverse spatial profiles of the two beams neglected in the theory may reduce the shift in the experiments. The relative jitter between the pump and probe pulses could also reduce the effect.

It is interesting to note in Fig. 2(c) that $\Delta T/T$ at a fixed wavelength can be either positive or negative depending on the carrier density. This is believed to be the first such observation. This phenomenon can be explained by a simple model assuming a Gaussian absorption line shape and further assuming the saturation of the oscillator strength as $f = f_0/(1 + N/N_s)$ and the broadening of the linewidth as $\gamma = \gamma_0(1 + N/N_x)$, where N is the carrier density and N_s and N_x are the characteristic carrier densities for the saturation and broadening. The model predicts that within a finite range of frequency detuning (from the center frequency) the sign of $\Delta\alpha$ depends on carrier density in agreement with the experimental observation. The frequency detuning for $\Delta\alpha = 0$ can be solved analytically and is N dependent.

Finally, one notes that in Fig. 2(g) that instead of a peak there exists a tiny dip at zero delay.[11] The tiny dip implies that the present model of a simple shift of the entire line shape is not complete. Instantaneous power broadening of the line shape due to the "virtual excitons",[10] which would explain this dip.

In conclusion, a blue shift due to the optical Stark effect was observed for the first time in InGaAs/InP MQW structures. The experimental results support the theory recently developed by Schmitt-Rink and Chemla and show that the scaling between the ternary and the binary III-V compound semiconductors is correct as concluded in the previous experiments.[7,12] This fast effect is potentially useful for subpicosecond optical switching.[13] Its existence at 1.5 μm makes it particularly attractive for switching in optical communication network.

[1]A. Mysyrowicz, D. Hulin, A. Antonetti, A. Migus, W. T. Masselink, and H. Morkoç, Phys. Rev. Lett. 56, 2748 (1986).
[2]A. Von Lehmen, D. S. Chemla, J. E. Zucker, and J. P. Heritage, Opt. Lett. 11, 609 (1986).
[3]W. T. Tsang, Appl. Phys. Lett. 45, 1234 (1984).
[4]InP is transparant in the spectral region of interest. Therefore, in the experiments the InP substrates were only slightly polished but not removed.
[5]The linewidth was deduced from the long-wavelength portion of the peak.
[6]W. T. Tsang and E. F. Schubert, Appl. Phys. Lett. 49, 220 (1986).
[7]K. Tai, J. Hegarty, and W. T. Tsang, Appl. Phys. Lett. 51, 86 (1987).
[8]A. Von Lehmen, J. E. Zucker, J. P. Heritage, D. S. Chemla, and A. C. Gossard, Appl. Phys. Lett. 48, 1479 (1986).
[9]This value was underestimated, because the exciton line was partially saturated and broadened by the carriers excited by the pump and probe beams (see Ref. 7).
[10]S. Schmitt-Rink and D. S. Chemla, Phys. Rev. Lett. 57, 2752 (1986).
[11]The peak disappears at 1508 nm followed by the emergence of the tiny dip at 1510 nm [Fig. 2(g)]. All the signals decay asymptotically to zero at longer wavelengths.
[12]K. Tai, J. L. Jewell, W. T. Tsang, H. Temkin, M. B. Panish, and Y. Twu, Appl. Phys. Lett. 50, 795 (1987).
[13]D. Hulin, A. Mysyrowicz, A. Antonetti, A. Migus, W. T. Masselink, H. Morkoç, H. M. Gibbs, and N. Peyghambarian, Appl. Phys. Lett. 49, 749 (1986).