Subpicosecond excitonic electroabsorption in room-temperature quantum wells

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We investigate the dynamics of excitonic optical absorption in room-temperature GaAs quantum wells during the application of a rapidly changing electric field in the plane of the quantum well layers. We obtain electroabsorptive modulation with a response time constant of 330 fs, which is the fastest ever reported in a semiconductor.

The optical absorption of semiconductors near the band gap is sensitive to applied electric field. In the Franz–Keldysh (F–K) effect[1,2] for example, the optical absorption below the band gap increases with increasing field. Exciton absorption resonances can be strongly perturbed usually at lower field strengths than commonly used to observe the F–K effect.[3] Although the F–K electroabsorption has been applied to optical modulators,[2] excitonic effects have not. Recently, quantum well structures consisting of alternating thin layers of two semiconductors such as GaAs and GaAlAs,[4] InGaAs and InAlAs,[4,5] and InGaAs and InP[6] have shown clear excitonic resonances at room temperature, and the excitonic electroabsorption has recently been investigated in the GaAs/GaAlAs system.[7,8] Since only small volumes are necessary for large optical modulation with these effects (e.g., a few cubic microns), devices utilizing these phenomena are particularly attractive for ultrafast optoelectronic applications. Problems such as velocity mismatch of optical and electrical waves are minimized, and because of the excitons sensitivity to applied fields increased detection sensitivity may be obtained in subpicosecond electrical sampling applications. In this letter we report the first subpicosecond investigation of excitonic electroabsorption. We have measured the response of the optical absorption of GaAs/GaAlAs quantum wells to electric field transients applied in the plane of the quantum well layers. The qualitatively different quantum-confined Stark effect, which is obtained when the field is applied perpendicular to the layers,[7,8] will not be considered here.

In bulk materials the exciton line(s) broaden with applied field because of the reduction of lifetime of the exciton by field ionization.[9] When fields of the order of the exciton classical ionization field are applied, the line broadening can be as large as the exciton binding energy and the resonance is completely smeared out. In this case the exciton is ionized in a time comparable to the classical orbit time. This effect can be treated in a unified theory with the F–K effect both for three- (Refs. 3 and 9) and two-dimensional[10] semiconductors with qualitatively similar results in both cases provided that the electric field is applied in the plane in the two-dimensional case. Weak excitonic electroabsorption (i.e., small change in transmission per applied field) can be observed at room temperature in bulk semiconductors[11]; however, in quantum wells the effect is large at room temperature with fields applied in the plane of the quantum wells.[8] The F–K effect has been shown to have subpicosecond response in direct time-resolved measurements in GaAs.[12] In a previous experiment the time response of the effect of parallel fields on excitonic absorption in quantum wells has been measured using a photoconductive switch to provide the electrical drive.[13] This investigation showed rise and fall of the optical transmission in 30 ps, presumed to be limited by geometrical effects in the sample electrode design. This work[13] is the fastest previous measurement of excitonic electroabsorption of which we are aware in any semiconductor.

The sample for the present measurements, which has been described in detail elsewhere,[8] consists of 60 GaAs quantum well layers, each 95 Å thick, separated by 98-Å barrier layers of $Ga_{0.68}Al_{0.32}As$ grown on top of a 2-μm transparent superlattice buffer and etch stop layer. The whole structure is grown on a GaAs substrate by molecular beam epitaxy. Contacts are formed into the top of the quantum well material region as shown in Fig. 1, the sample is epoxied to sapphire, and the GaAs substrate is removed in the vicinity of the electrodes.[8] A voltage pulse of 5 μs duration applied between the electrodes results in an applied electric field in the plane of the quantum wells. All measurements are performed at room temperature.

Optical pulses of 50 fs duration are produced by a colliding-pulse mode-locked laser[14] with internal Brewster prisms for group-velocity compensation.[15] The pulses are amplified by a copper vapor laser pumped amplifier[16] that produces single amplified pulses of 4 μJ energy at 8 kHz repetition rate. These pulses are focused to near the diffraction limit into a 1-mm jet of ethylene glycol, thus generating white light continuum pulses which extend into the ultraviolet and to at least 900 nm in the infrared. A portion of the infrared continuum centered at the heavy-hole exciton energy ($\lambda = 855$ nm) is selected with a 10-nm bandwidth interference filter for use as the probe pulse. This spectral selection increases the probe pulse width to ~ 120 fs but prevents substantial excitonic absorption saturation by unwanted probe light at other wavelengths. A section of the continuum around 700 nm is selected with glass cutoff filters for use as a pump pulse. The time response limit set by the optical pulse durations was measured to be ~ 130 fs by time resolving the

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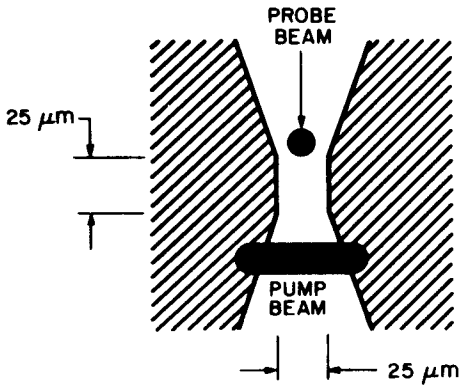

FIG. 1. Schematic diagram of the electrode structure on the surface of the quantum well sample. The positions and sizes of the pump and probe spots are also shown approximately to scale.

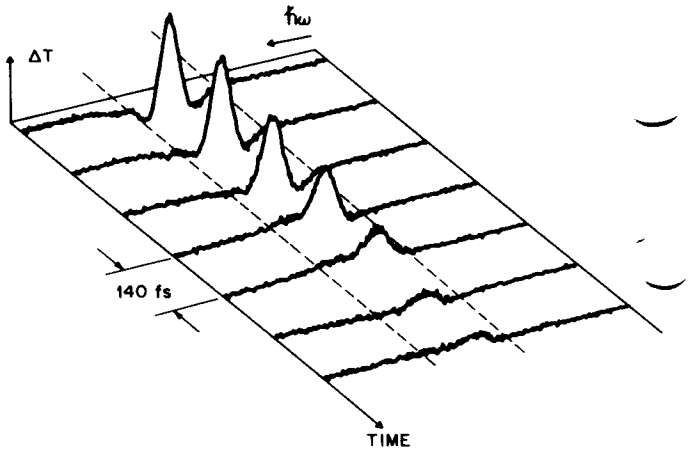

FIG. 2. Electric field differential spectra as a function of probe beam del
The bandwidth of the filter in the incident probe beam is indicated by t
dashed lines.

excitonic saturation response with pump and probe beams overlapped on the sample.

To generate a rapid change in field, the pump pulse is deposited on the sample (Fig. 1), uniformly illuminating the space between the electrodes on one side of the gap. This creates a high-density electron-hole plasma ($\sim 10^{19}$ cm^{-3}) that shorts out the field between the electrodes. The probe pulse passes through the sample on the other side of the gap with a variable time delay derived from a stepper-motor-controlled delay line and measures the response of the excitons to the applied electric field.

This particular electrode structure (unlike, for example, the coplanar stripline[17]) is not well suited to studies of generation and propagation of electromagnetic transients in the THz regime. Consequently, to minimize propagation and dispersive effects, we measure the excitonic response very close to the point at which the field transient is generated ($\sim 50\,\mu$m). The recovery of the field is expected to occur on a time scale which is much longer than that considered here and will involve many reflections from the electrode structure as well as external propagation effects from the bias connections.

The transmitted probe pulse is detected with a Reticon optical multichannel analyzer (PAR OMA-III) and spectrometer operating in a new differential scanning mode. In this mode, the OMA accumulates one scan (50 ms), during which time the electrical bias pulses (35 V) are applied to the sample. Four hundred spectra are integrated in this scan. Next, the bias pulses are inhibited, the OMA accumulates another scan, and subtracts this from the previous scan. This process is repeated 500 times in this experiment. The data are then stored, the delay line is advanced, and the process is repeated.

When the pump pulse arrives after the probe, as in the earliest spectra in Fig. 2, this procedure generates an electric field differential spectrum, as in Ref. 8, albeit truncated by the filter spectral bandwidth. At the position of the heavy-hole exciton resonance peak, the transmission increases because of the broadening of the exciton resonance with field, while that at lower and higher energies decreases for the same reason. However, as we delay the arrival of the probe pulse, we can see in Fig. 2 that the magnitude of the electric field differential spectrum decreases. There is also some evidence of a small change in the form in the spectra at later times, with the dip at short wavelengths disappearing.

We interpret the changes in the spectra as being the response of the excitonic optical absorption to a rapidly changing electric field. The time behavior of the differential absorption at the heavy-hole peak is shown in Fig. 3. These results are obtained by averaging the differential spectra over the exciton full width at half-maximum. The transmission change falls off with a time constant of $\sim 330 \pm 50$ f giving a 90–10% fall time of 660 $\pm$ 100 fs. Moving the pump and probe spots further apart gave longer time constants (e.g., 1.6 ps 90–10% fall time at 100-μm spot spacing) suggesting that the time response is dominated by electromagnetic propagation effects (such as reflections and dispersion) in this nonideal transmission geometry. It is interesting to note that these times are approaching the classical orbit time of the exciton which is ~ 150 fs for an exciton of 10 meV binding energy. We might expect this to set a

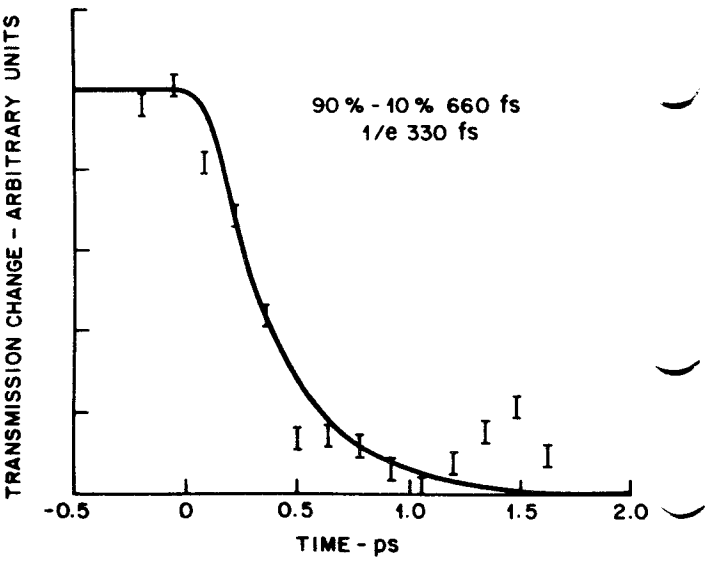

FIG. 3. Amplitude of the differential spectrum at the heavy-hole peak as a function of probe delay.

fundamental limit to the electroabsorption response time; however, at present the measured differential spectra do not indicate any significant dynamic shape changes which may be indicative of an approach to a fundamental limit. One other process that could contribute to very fast changes in absorption in the present experiment is saturation of the excitonic absorption of the probe by the pump beam[18]; however, this is not significant in the present experiment since the pump intensity at the position of the probe beam is considerably lower than the probe beam intensity itself, and the probe beam intensity is made small enough that it causes very little self saturation.

Finally, since the results in this letter show fast response of the excitonic electroabsorption, it is of interest to consider the potential sensitivity of this effect for measurement of femtosecond electrical transients. We can estimate dT/dV (T is the transmission), an important parameter in comparing sensitivities, giving $dT/dV \sim 0.01\ V^{-1}$ at 35 V. In comparison, the high-speed measurement system demonstrated by Valdmanis *et al.*[19] is based on a device sensitivity of $\sim 8\times 10^{-4}\ V^{-1}$. This suggests that the present quantum well material, if incorporated in a similar system, might be capable of achieving even higher sensitivities. Furthermore, narrower electrode spacings (e.g., 10 μm) may be used for correspondingly higher sensitivities. A high-repetition-rate source comparable to the colliding pulse mode-locked laser but operating at ~ 855 nm would, however, be required to exploit these advantages fully.

In conclusion, we have demonstrated subpicosecond excitonic electroabsorption for the first time. We have shown that this is also potentially a very sensitive phenomenon, applicable to high-speed electro-optic sampling with very small device sizes. Although demonstrated with a quantum well structure, which enables room-temperature operation, the same phenomenon should be observable in other, cooled semiconductor materials, and also in other quantum well material systems. The speed of response measured here does not yet appear to be a fundamental limit, and is ascribed instead to geometrical factors in the electrode structure. The physics of these limits in improved structures will be the subject of further work.

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