Femtosecond ac Stark Effect in Semiconductor Quantum Wells: Extreme Low- and High-Intensity Limits

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We show that in quasi-2D GaAs quantum wells, below-resonance low-intensity excitation produces a *pure shift* of the excitons, contrary to ac Stark shifts in atomic systems. At high pump intensities two-photon 3D-real-carrier generation competes with the virtual 2D-exciton effects. We also show that dc fields applied perpendicular to the layers reduce the excitonic ac Stark effect.

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It has been shown recently that dynamical Stark shifts of the band-edge states are produced by intense photoexcitation of semiconductors below the band gap, E_g.[1,2] Because of the extended nature and strong mutual interaction of the electronic excitations in semiconductors, the excitonic ac Stark effect (ac SE) is more complicated in character[3,4] than that in atomic systems.[5] Rather, the theory[3,4] reveals profound similarities between the photogenerated populations of virtual excitons and Bose condensed gases (at small or moderate detuning and intensity) or superconductors (at large detuning and intensity). A marked difference between atomic and excitonic ac SE has been recently predicted by numerical calculations[6-8] based on the theory of Refs. 3 and 4. It has been shown that in contrast to atomic systems, for which the ac SE is always accompanied by a reduction of oscillator strength, in two-dimensional (2D) semiconductor quantum wells (QW) low-intensity, moderate-detuning, below-gap excitation should produce a *pure shift* of the exciton resonances. It is only at high intensity that the resonances lose height and broaden.[6]

The excitonic optical nonlinearities originate from anharmonicities in the exciton-exciton and exciton-photon interaction which depend directly on the polarization and density of virtual populations.[3,4] In turn, the distribution of the latter over the bound and scattering exciton states is determined by the intensity I_p and detuning $E_g - \hbar\omega_p$ of the pump laser.[8] Thus, experimental investigations of the excitonic absorption under various conditions of below-gap excitation provide information on a novel "condensed" many-body system.[3,4,6-8] The signature of virtual populations is that they last only as long as the optical excitation; therefore they are studied by time-resolved techniques using ultrashort laser pulses.[1,2] We have found that, at the large detunings required for ultrashort (< 100 fs) excitation, a systematic study of the effects of virtual populations without contributions from real populations is complicated in practice by nonresonant two-photon carrier generation. This occurs because the ac Stark shift decreases approximately inversely in proportion to the detuning[2-4] whereas the two-photon absorption (TPA) cross section is relatively constant.[9] Hence TPA becomes increasingly important relative to the ac SE as the detuning increases.

In the case of QW's, the TPA real carriers are essentially 3D because they are generated at energies of $\approx 2E_g$, i.e., well above the gap discontinuity between the QW and the barrier-layer material. Their interaction with the 2D band-edge excitons is thus completely different from that of the 2D virtual excitons. In QW's the phase space filling (PSF) of 2D excitons has a strong contribution to the ac SE while corresponding 2D exciton screening is small.[10,11] On the contrary, the high-energy 3D carriers do not contribute to PSF but do screen the electron-hole interaction dynamically, giving rise to collision broadening. Through the resulting coexistence of virtual 2D excitons and real 3D carriers novel and interesting aspects of dimensionality can be explored.

In this Letter we report investigations of the effects of virtual excitons and high-energy real carriers on the band-edge absorption of GaAs QW's. We use a special sample which consists of a *p-i-n* diode with 74-Å QW in the intrinsic region. By high-sensitivity femtosecond spectroscopy we can sort out the respective contributions of the two species on the absorption spectra, and by measuring the intensity dependence of the photocurrent in the *p-i-n* diode we can unambiguously assign the origin of the real carriers to TPA. We demonstrate that at very low excitation intensity and moderate detuning the virtual excitons induce *a pure shift* of the band-edge excitons without loss of oscillator strength, in agreement with Refs. 6–8. At higher intensity both a shift and a decrease in exciton absorption due to virtual populations are observed.[1] At very large pump intensities we find that TPA is strong and unavoidable and is a natural

consequence of excitation below the band edge with ultrashort optical pulses. We follow the evolution of the exciton differential line shape versus intensity and we compare it to that induced by real-carrier populations excited by TPA.

For our experiments we use a newly developed laser system which generates 100-fs optical pulses of microjoule energies at a 8-kHz repetition rate with center wavelength at 805 nm.[12] Pump pulses of 100-fs duration are selected from an intense infrared continuum which is generated in a jet of ethylene glycol with interference filters around 820 nm. We measure differential transmission spectra with an optical multichannel analyzer and spectrometer operating in differential detection mode while synchronously chopping the pump beam. We scan the time delay between the pump and probe beams to obtain the full time course. Kilohertz repetition rates allow us to use differential detection techniques for small-signal analysis. The 74-Å-QW exciton absorption line is at 780 nm. Since the femtosecond white-light continuum center is 805 nm, we now have available extremely intense (up to 1 TW cm^{-2}) continuously tunable excitation pulses below the band gap of GaAs quantum wells.

We first performed low-excitation-intensity experiments. In Fig. 1(d) we show the absorption spectrum $\alpha(\omega)$ of the sample at 35 K measured with very-low-intensity ($I_l \approx 10$ kW cm^{-2}), 100-fs continuum pulses. When the sample is excited ≈ 50 meV below the heavy-hole exciton with low-intensity 100-fs pump pulses ($I_p \approx 30$ MW cm^{-2}), the changes of the absorption spectrum are very small, only a few percent, i.e., about the width of the pen line in Fig. 1(d). They can be seen only by measuring the differential transmission spectrum (DTS) $\Delta T/T \approx \Delta\alpha \times l$. In the limit of a pure shift, the DTS should have exactly the same line shape as the derivative of the linear absorption $\partial\alpha/\partial\omega$. In Fig. 1(a), $\partial\alpha/\partial\omega$ calculated from $\alpha(\omega)$ of Fig. 1(d) is compared to the measured DTS at zero time delay. We find that around the exciton resonance the two line shapes are identical, in agreement with the theory.[3,4,6-8] The origin of a pure shift of the exciton without loss of oscillator strength can be explained as follows. The virtual excitons generated by the below-resonance excitation produce a reduction of the oscillator strength owing to PSF and a shift of the bound states *as well as of the scattering states*,[6-8] i.e., the continuum of unbound excitons. At moderate detuning, however, the shift of the band edge is larger than that of the $1S$ exciton,[6,7] so that the "binding energy" of the exciton in the presence of the virtual populations is *increased*. Since the exciton oscillator strength is proportional to its binding energy, its oscillator strength increases. In the case of 2D systems at low excitation, this enhancement cancels almost exactly the loss of exciton oscillator strength due to virtual-exciton PSF.[6-8] Physically, this effect reflects once more the larger interaction cross section with a given exciton

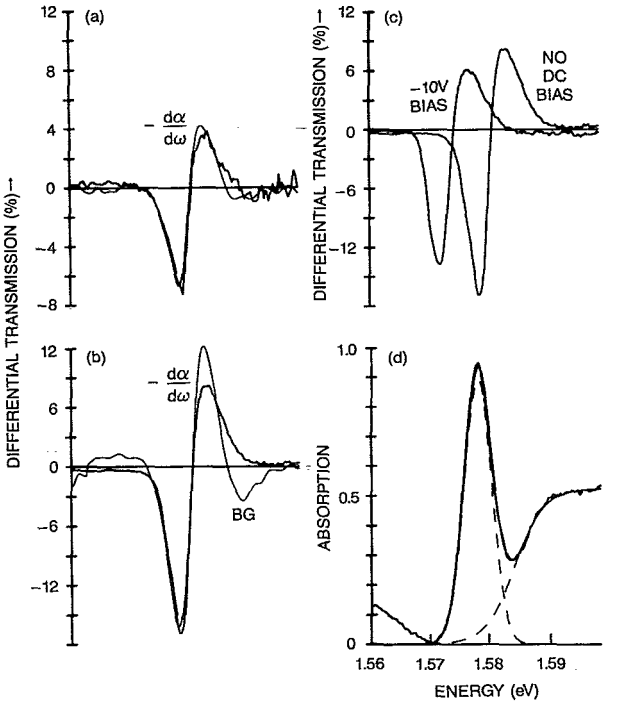

FIG. 1. (a) Differential transmission spectrum at delay $t=0$ obtained at a pump intensity $I_p \approx 30$ MW cm^{-2}. The derivative of the experimental absorption (d) with respect to frequency is also shown, demonstrating that the exciton exhibits a pure shift at low intensities. (b) Differential transmission spectrum at $I_p \approx 100$ MW cm^{-2}. The signal no longer matches the derivative of the sample absorption, indicating a departure from pure shift of the exciton. The negative dip denoted by BG on the derivative of the linear absorption indicates the signal that is expected in the case of a simple rigid shift of the band-edge states. (c) Differential transmission spectrum at $\Delta t=0$ for a 50-meV detuning at 0 V applied dc bias, and for the same pump wavelength and intensity, but with -10 V dc bias applied to the sample. The ac SE is *reduced* in the presence of a $F \approx 10^5$ V/cm dc bias field perpendicular to the quantum wells. (d) Absorption spectrum of the $Al_{0.3}Ga_{0.7}As$/GaAs *p-i-n* diode containing fifty 74-Å GaAs quantum wells in the intrinsic region at 0 V bias. Dashed lines show the components of a simple fit to the absorption spectrum using a Gaussian exciton resonance and a sigmoid continuum function. All the data presented in this article were obtained at 35 K.

of the scattering states compared to that of bound states. Only for very large detuning does the $1S$ exciton state and the continuum shift by the same amount.[8] Although the exciton binding energy increases to keep the excitonic absorption area constant, detailed theoretical calculations of the total spectrum suggest that there should not be a simple feature in the spectrum characteristic of a simple shift and bleaching of the continuum absorption edge, in qualitative agreement with our measurement. We note, however, that recently Joffre, Fluegel, and Peyghambarian[13] have reported continuum shifts in the 3D bulk CdS, at much larger detuning.

Experiments with *weak pump* but at *higher test inten-*

sities indicate a departure from this pure-shift regime, presumably because of a nonnegligible population of real excitons created directly by the test beam. We have found that only for $I_t < 10$ kW cm^{-2} is the effect of the test beam negligible, and all our experiments were performed under these conditions. When the *pump intensity* is increased one observes evidence of both a shift and a decrease in the exciton absorption, as predicted theoretically[6] and seen earlier.[1] This translates in the DTS to a decrease and broadening of the positive lobe [see Fig. 1(b)]. Nevertheless, these effects are still due to virtual populations since they last only as long as the pump pulse is applied and the absorption completely recovers at $t > 150$ fs.

Yamanishi and co-workers[14] have theoretically investigated the ac SE under dc applied fields perpendicular to the QW's. The effect of such an applied field is to shift the excitonic resonances to lower energies, due to the quantum confined Stark effect.[15] With a detuning of 50 meV at zero bias, we find that the applied dc field *reduces* the ac SE [Fig. 1(c)], even though the quantum confined Stark effect reduces the detuning by ≈ 6 meV. The rapid decrease of electron-hole overlap with applied field dominates in this case.

Further increase in the pump intensity brings about a more complicated regime which has not been systematically investigated to date. This is shown in Fig. 2 where we present the full time course of the DTS obtained with 100-fs excitation pulses at 80 meV below the exciton energy and $I_p \approx 3$ GW cm^{-2}. This plot is obtained by overlaying a grid on top of our ten measured DTS. We observe a rapid initial transient at both the heavy-hole and light-hole excitons. This transient contains both an excitonic shift and bleaching. After the rapid transient the signals do not recover to zero. On our time scale the DTS shows a very long-lived component (many ps). The heavy-hole exciton DTS at +264 fs is shown as curve *a* in the inset of Fig. 2. The ac SE does not contribute to the signal at this time delay since the pump pulse is only 100 fs long, and we find that this signal is the signature of a pure exciton line broadening. The integral of the $t = +264$ fs transmission change over the heavy-hole exciton line is also shown in the inset of Fig. 2, as curve *b*. The integrated transmission change around the exciton resonance is nearly zero, which results since there is no direct occupation of band-edge states.[10,11] This signal appears to be due to a dynamical screening of the electron-hole interaction such as that which a real plasma would produce. Such a plasma can be generated by TPA.[9]

To investigate the presence and origin of real carriers we have studied the photocurrent in a *p-i-n* diode with a reverse bias of 10 V. We find indeed a photocurrent with a clear quadratic dependence on I_p as shown in Fig. 3. We estimate, using the known TPA coefficient of GaAs,[9] that at 3 GW cm^{-2} we expect a photocurrent of 2 μA, about 10 times higher than we measure. The photoconduction quantum efficiency in perpendicular trans-

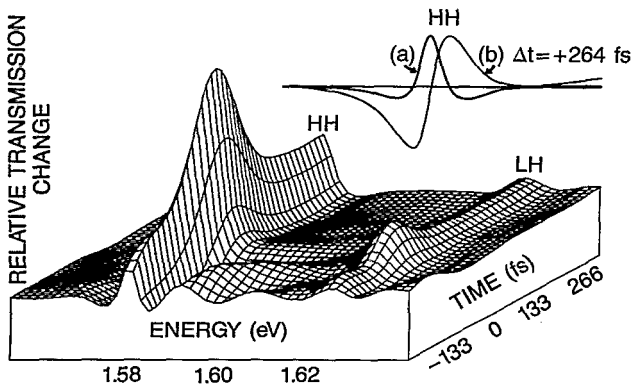

FIG. 2. Time course of differential spectrum for excitation 80 meV below the heavy-hole exciton at $I_p \approx 3$ GW cm^{-2}. Signals at the heavy-hole (HH) and light-hole (LH) exciton are simultaneously observed. The nonzero signal at late times (> 264 fs) is due to two-photon carrier generation. Inset: curve *a* shows the differential transmission spectrum around the HH exciton, and its integral, curve *b*, demonstrates the area conservation resulting from screening of the 2D excitons by the 3D plasma.

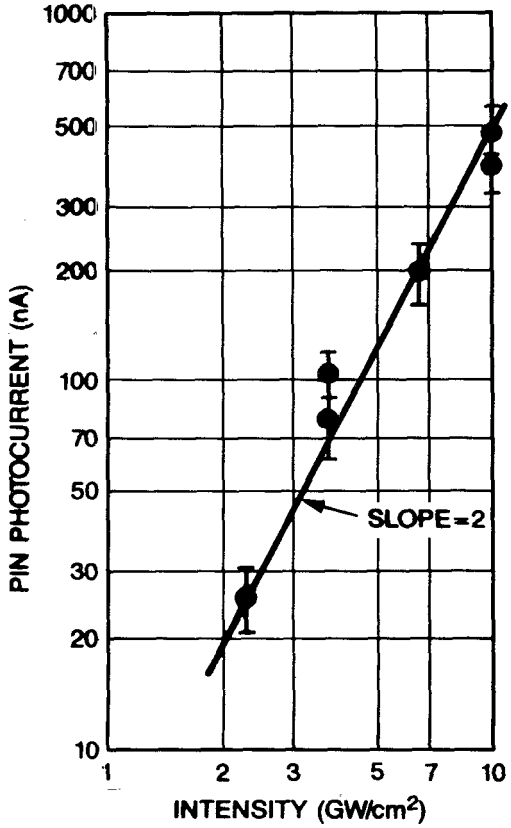

FIG. 3. Photocurrent measured with 10 V reverse bias applied on the *p-i-n* quantum-well sample, showing a quadratic dependence on the pump intensity.

port at low temperature in quantum wells is not well understood; however, our estimates of the expected TPA photocurrent are in reasonable agreement with our measured values. The carriers excited by TPA are 3D in character since their excess energy (≈ 1.5 eV) is far above the confinement potential in our QW (200 meV). They are free to move in 3D and able to screen instantaneously the 2D excitons effectively. This effect prevents the full recovery of the excitonic absorption. The perturbation caused by these carriers becomes stronger in time as they relax down to the band edge.[16,17] Then they will occupy $k=0$ states and bleach the exciton line in such a way that the integrated transmission change will no longer be zero. Of course, after the carriers recombine in a few ns the excitonic absorption fully recovers. We note that spectral oscillations at negative time delays are clearly observed in these data. These are due to the interaction of the pump pulse with the polarization induced by the probe field.[18]

In conclusion, we have shown that below-gap excitation in quasi-2D GaAs QW's produces, at low pump intensity, a *pure shift* of the exciton resonances, unlike two-level atoms. As the pump intensity increases both a shift and a decrease in absorption of the excitons due to virtual populations are observed. At very large pump intensities and large detuning two-photon carrier generation becomes important. This causes power dissipation and long (ns) lifetime transients and is undesirable for applications. Use of a reverse-biased *p-i-n* structure provides an independent measure of the two-photon-generated photocarrier density. In the small-detuning limit, shifts of the excitons can be obtained at lower pump powers, so that real-carrier two-photon effects are weaker; however, very short optical pulses (less than 100 fs), because of their large bandwidth, inherently violate the small-detuning limit. Virtual excitation and two-photon carrier excitation effects are connected in a fundamental way and a full theoretical analysis of their inter-relation is required.

Part of this work was previously reported.[19]

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Note added.—The simultaneous occurrence of ac SE and TPA was also observed in atomic vapors.[20]

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