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Virtual excitation of the Fermi-edge singularity in modulation-doped quantum wells

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The Fermi-edge singularity (FES) in modulation-doped GaAs quantum wells is a unique many-body feature that arises from strong Coulomb correlations within a dense sea of electrons. Using intense ultrashort excitation, we show experimentally and theoretically that the nonresonant nonlinearity (ac Stark shift) of the FES is dramatically different than that of the excitonic ac Stark shift of undoped-quantum-well excitons.

A nonresonant optical field can interact with electronic transitions to produce significant changes in the absorption spectra. This effect has been called the optical or ac Stark effect (ACSE) and it is manifested as a spectral shift of the optical transitions usually followed by a loss of their oscillator strength, both occurring while the optical field is present in the sample. This effect has been observed in atoms;[1] however, the existence of collective electronic excitations makes the ACSE in semiconductors different.[2] It was shown that excitonic effects can dramatically alter the bleaching that is routinely observed in atomic systems. Excitons in undoped quantum wells (QW's) have been extensively studied with below-band-gap excitation using ultrashort pulses.[3,4] While excitonic transitions exhibit the expected instantaneous blueshift, the two-dimensional continuum does not exhibit a shift.[4]

So far, all the experimental and theoretical work on the excitonic optical Stark effect in semiconductors has been carried out in undoped samples. An interesting and unexplored case is that of modulation-doped QW's (MDQW's), in which the presence of the electron Fermi sea dramatically changes the nature of the optical properties in a manner similar to the x-ray singularity in metals originally considered by Mahan.[5] In these samples the absorption edge at low temperatures is governed by the Fermi-edge singularity (FES), which results from the many-body correlations between the Fermi sea and the photoexcited hole. Although the FES can result in a sharp spectral feature,[6] it is a continuum many-body resonance and has no binding energy. Theoretically, models of the FES are incomplete because of a number of complicating factors, such as finite mass, band-structure effects, and dynamical properties of the Fermi sea. Nonresonant optical excitations present another dimension to the study of many-body correlations in the high-density regime. In this paper we present the experimental observation of ac Stark shifts of the FES and a theoretical description that accounts for the observed behavior. For small intensities, we find that the FES shifts to the blue by an amount comparable to that observed in undoped QW's under the same excitation conditions. For higher intensities though, the FES does not show the considerable bleaching that is usually observed either in atomic systems or in other undoped semiconductors. We demonstrate how the distinct origin of the resonance in doped and undoped samples leads to the different transient behavior. We present numerical results obtained from the quantum-kinetic equations that demonstrate the importance of screening dynamics within the Fermi sea and agree qualitatively with the experimental observations.

We study GaAs/$Al_{0.3}Ga_{0.7}As$ MDQW samples of various widths and with carrier concentrations of $n=(1-3)\times10^{11}$ cm^{-2}. In this paper we will concentrate on the sample that shows the sharpest FES, although similar results were obtained for the rest of the samples. This sample has 12-nm-wide wells separated by 35-nm barriers. The central 12 nm of the barriers are Si doped providing an electron concentration of 2.6×10^{11} cm^{-2}, yielding a Fermi energy of $\sim$15 meV at 4 K. The absorption peak at $\sim$1.54 eV corresponds to the FES (Fig. 1). The FES exhibits a strong temperature dependence in the range 8–40 K, as expected for a true FES. We note that the other features are due to higher subband transitions, and they remain basically unchanged even for temperatures close to 100 K. The corresponding undoped sample has 10-nm wells and 10-nm $Al_{0.3}Ga_{0.7}As$ barriers. The ac Stark effect is measured by recording the femtosecond transient changes in absorption induced by an intense

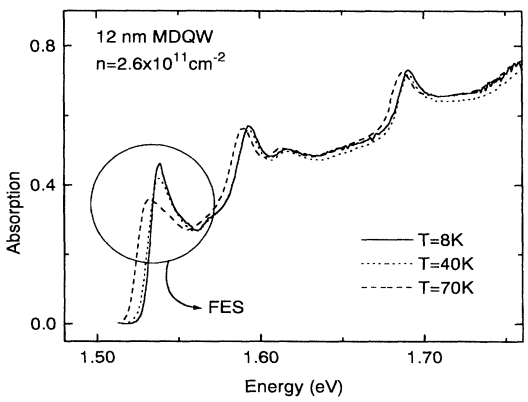

FIG. 1. Absorption spectrum of the n-type modulation-doped quantum-well sample at three temperatures, showing the Fermi-edge singularity (FES) at 1.54 eV. The sharp temperature dependence is a characteristic of the FES. Other higher-energy interband transitions are unaffected in this temperature range.

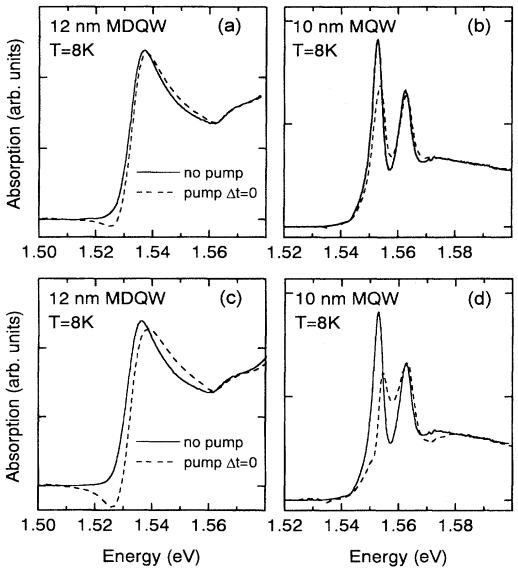

FIG. 2. Optical-absorption spectra measured with 100-fs pump and probe pulses. The pump is detuned 50 meV below the lowest transition in each case. (a) n-type sample at ~ 1 GW/cm^2; (b) undoped sample at ~ 1 GW/cm^2; (c) n-type sample at ~ 5 GW/cm^2; (d) undoped sample at ~ 5 GW/cm^2. In each case, the dashed line shows the absorption spectrum in the presence of the pump beam, and the solid line shows the linear-absorption spectrum (no pump).

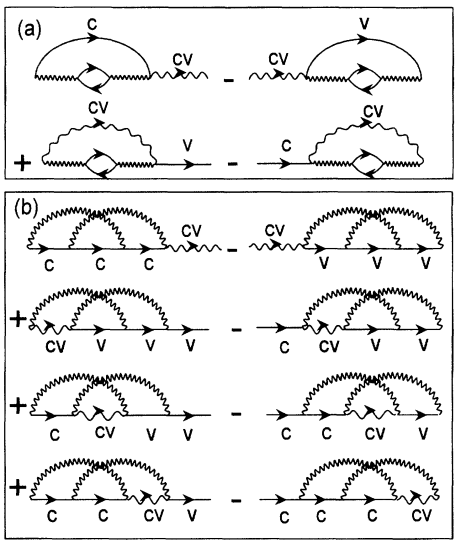

FIG. 3. Second-order self-energy and vertex corrections contributing to the dephasing of the polarization. (a) Screened Hartree-Fock contribution and (b) corresponding exchange contributions. Contributions due to polarization scattering are not shown.

below-band-gap excitation pulse. Pulses of 60-fs duration are generated in a self-mode-locked Ti-sapphire laser and amplified to microjoule energies at an 8-kHz repetition rate in a copper-vapor laser-pumped dye amplifier. The pulses are then focused in an ethylene-glycol jet to generate white-light continuum pulses. For the probe beam, we use a small fraction of the continuum (less than 1%) with its full bandwidth. The pump beam is intense (1–5 GW/cm^2) and its spectrum is selected with an interference filter of 12-nm bandwidth. Quarter-wave plates were placed in both beams to allow for circular polarization control. The samples were mounted in a He-flow cryostat and in all experiments the samples were maintained at a temperature of 8 K.

We compare the absorption spectrum with and without the pump pulse exciting the sample, and we compare the undoped and doped samples at nearly identical detuning and intensity. Figures 2(a) and 2(b) show the absorption of doped and undoped samples, respectively, at an excitation intensity of ~ 1 GW/cm^2. The solid curves show the absorption spectra with the pump off, and the dashed lines show the ac Stark-shifted absorption curves for a nominal zero time delay between pump and probe beams. The detunings of the pump was ~ 50 meV for both samples, and both pump and probe beams were σ^+ polarized. In the MDQW we measure the detuning with respect to the FES. Figures 2(c) and 2(d) show the same sets of data for higher pump intensities (~ 5 GW/cm^2). We point out three principal observations. First, under nearly identical conditions of intensity and detuning, the blueshift of the FES is practically the same as that of the exciton in undoped samples: (~ 1 meV at 1 GW/cm^2). Second, the FES exhibits very little loss of peak height as it shifts to higher energy. In comparison, the heavy-hole exciton transition in the undoped sample exhibits a significant bleaching at both low and high intensities. These observations, namely, the similarity of the blueshift for corresponding doped and undoped samples and the lack of bleaching in the doped ones, *were not dependent on detuning, intensity nor doping density* (in the appropriate range where the FES can still be observed). Also, the *same observations* were measured in other sample pairs with *different well widths*.

From a theoretical point of view, the FES is a complex many-body resonance.[5] In order to properly handle the time dependence in this problem, we have to solve the semiconductor Bloch equations,[7] accounting for the dynamical response of the Fermi sea microscopically. As has been shown,[7] simple light-induced gap-renormalization arguments do not adequately describe the complex situation due to the existence of exciton-exciton interactions, Pauli blocking, and the spectral width of the pulse. In addition, to properly describe the doped case (FES), we find that it is necessary to account for additional light-induced carrier-dephasing mechanisms that are not present in undoped semiconductors. These scattering mechanisms are different from those that determine the FES line shape. We start from the general quantum-kinetic equations for diagonal and off-diagonal elements of the one-particle propagators.[8] Self-energy and vertex corrections arising from interband contributions to the self-energy are treated on an equal footing within a second-order Born approximation. Corresponding contributions to the dephasing of the polarization are shown diagrammatically in Fig. 3. We include third-order polarization-scattering contributions, which are of the same order of magnitude in the coherent regime. Corresponding particle and polarization-scattering contributions occur in the equation for the distribution functions of electrons and holes. We perform a usual Markovian approximation for these scattering

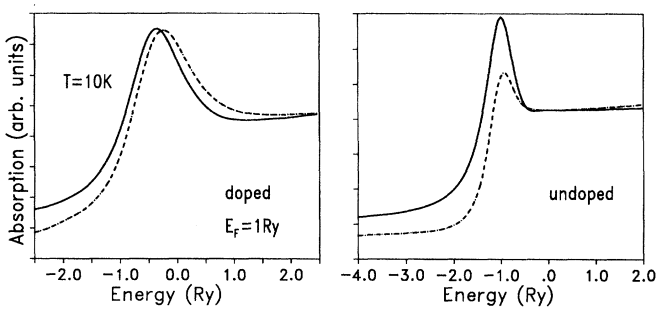

FIG. 4. Results of numerical calculation for (a) n-type doped sample with Fermi energy of 1 Ry and a pump detuning of 6 Ry, and (b) undoped sample under same conditions as (a).

contributions entering the semiconductor Bloch equations. Apart from the contributions corresponding to exchange-type self-energies or vertex corrections [Fig. 3(b)], respectively, analytical results for the Hartree-Fock dephasing contribution [Fig. 3(a)] have been given in Ref. 9 for the case of a screened Coulomb potential. Thus in our description the unbinding of the Mahan exciton results not from the balance between ladder and crossing diagrams but from the screening of the Coulomb interaction, if the latter is treated realistically instead of introducing a contact interaction. There are several differences in the present approach in comparison to other theories. Most theoretical approaches to the FES problem[10] approximate the screened Coulomb interaction by a contact potential. With this approximation, there is a systematic compensation between nth-order ladder diagrams and corresponding diagrams with maximal crossing. It is just this balance between ladder diagrams (e-h attraction) and crossing diagrams (excitation of the Fermi sea due to the action of the valence hole) that converts the exciton resonance into a power-law singularity. This has also been discussed by Bauer.[11] This behavior changes if a more realistic screened Coulomb interaction is considered. For a contact interaction the ratio of the second-order ladder and crossed diagrams is $\frac{1}{3}$. In the case of a statically screened Coulomb interaction, this ratio becomes density dependent and is roughly $\frac{1}{8}$, when $r_s \sim 1$. This indicates that the contribution of maximal crossed diagrams compared to ladder diagrams is considerably reduced for the case of a screened Coulomb potential. Therefore, we neglect diagrams with more than two crossing Coulomb lines. A second feature of the present approach is the consideration of finite hole mass, which has been shown to smear the edge singularity over an energy range of the order of the hole recoil energy.[12] A quantitative description of the underlying processes is only possible if e-e and e-h scattering is consistently taken into account. This is in contrast to most theories which have ignored the e-e interaction. Figure 4 shows the results of the numerical calculation carried out for the common pump-probe configuration[2] using bulk GaAs parameters. In the case of undoped materials, we use a phenomenological dephasing rate, as induced interaction effects are small in comparison with the intrinsic broadening. For the doped case, the line shape is determined by e-e interactions. In Fig. 4(a), we show the calculated absorption spectrum when pumping with a 100-fs pulse at 6 Ry below the band gap in the case of a Fermi energy of 1 Ry (=4.2 meV). In comparison, Fig. 4(b) shows the results for an undoped sample. The FES exhibits a blueshift that is comparable to that of the exciton in the undoped sample; however, the bleaching (loss of peak height) of the FES is much smaller than that of the exciton in the undoped sample. This agrees with our principal experimental observation. For the undoped case, it has been shown that the resulting bleaching can be attributed to the finite spectral width of the pump pulse,[7] and is missing in the stationary limit.[13] For the doped case, we find by numerical analysis that induced interaction effects are responsible for the lack of bleaching. The principal mechanism can be identified as the contributions of the random-phase approximation (RPA) self-energy to the dephasing. Dephasing is caused by scattering of the pump polarization $P_{\mathbf{k}}$ with occupied states of the Fermi sea $\mathbf{k}_1+\mathbf{k}_2$ into empty states $\mathbf{k}+\mathbf{k}_1$ and $\mathbf{k}_2$, respectively. The same processes contribute to the dephasing of the probe polarization. Contributions beyond RPA and from vertex corrections act in a similar manner and act to reduce the bleaching as well as the blueshift of the FES. This will be discussed further in a future publication. In comparison, broadening due to exciton-induced dephasing has been recently studied in GaAs.[14] Our light-induced dephasing processes in doped QW's are clearly of different physical origin.

In conclusion, we have shown experimentally and theoretically that the FES in doped QW's exhibits a very different ACSE behavior compared to the exciton in undoped quantum wells. We can account for this behavior with a numerical treatment of the quantum-kinetic equations. We find that the FES behaves in some aspects like an ordinary exciton, in that it exhibits a normal ac Stark shift. On the other hand, being a many-body correlation effect, it displays completely different transient behavior as compared to the undoped case. We find that the nonresonant nonlinear response of the FES is strongly modified by the scattering of carriers created by the probe beam with electrons in the Fermi sea. It is the difference in the actual *dephasing mechanisms* in doped and undoped samples that determines the observed difference in behavior, not just the magnitude of effective macroscopic dephasing times. In fact, the macroscopic dephasing times are not so different in both cases. The consequences of these different dephasing mechanisms on an ultrashort time scale have not been investigated up to now. Ultrashort pulse experiments provide a natural way to study complex scattering processes, and our hope is that they can improve our understanding of high-density dynamical phenomena in semiconductors.

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