Femtosecond Dynamics of Excitons under Extreme Magnetic Confinement

J. B. Stark,[a] W. H. Knox, and D. S. Chemla
AT&T Bell Laboratories, Holmdel, New Jersey 07733

W. Schäfer,[b] S. Schmitt-Rink, and C. Stafford[c]
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 10 July 1990)

The resonant optical nonlinearities of GaAs/AlGaAs quantum wells are measured with femtosecond time resolution, as the quasi-two-dimensional states are further confined into quasizero dimensions by a perpendicular magnetic field. We apply fields up to 12 T to demonstrate that exciton-exciton Coulomb interactions are strongly modified by quasi-zero-dimensional confinement, in agreement with many-body theory. These measurements demonstrate for the first time the remarkable result that at high magnetic fields an ensemble of 1*s* electron-hole pairs behaves like a gas of noninteracting particles.

PACS numbers: 78.20.Ls, 42.65.Re, 73.20.Dx

Using layered growth techniques to produce quasi-two-dimensional (2D) confinement of electronic states in quantum wells (QWs), it has been demonstrated that optical and electronic processes in semiconductors depend strongly on dimensionality. Quantum confinement significantly modifies fundamental properties such as the interparticle Coulomb interaction, the density of states, and the response to external fields. Excitons, the lowest-energy excitations in the intrinsic material, are ideal probes of the dimensional dependence of such properties: Their binding energy measures the strength of the Coulomb interaction; their response to internal or external fields determines their polarizability; and their nonlinear optical properties yield information on their interactions with each other, with free carriers, and with optical fields.[1] Further confinement within the QW planes can be obtained by applying a perpendicular magnetic field.[2] In this way, the dimensionality of the states in a QW structure can be continuously tuned from quasi-two to quasizero dimensions (0D), in uniform materials with excellent optical and electronic quality. In this Letter, we report the first femtosecond measurement of the nonlinear optical response of a magnetically confined 0D electron-hole system. The results of these studies provide information on the dimensional dependence of exciton-exciton and exciton-photon interactions, on an ultrafast time scale.

The relative motion of an optically excited 2D electron-hole pair in a perpendicular magnetic field H is determined by an effective potential, which is made up of two terms: The first term is the usual $-e^2/\varepsilon_0 r$ electron-hole Coulomb attraction; the second is a quadratic potential term $e^2H^2r^2/8mc^2$ imposed by the magnetic field ($\hbar=1$).[2] In the absence of the Coulomb interaction, the electron-hole eigenstates would be the energetically discrete and equally spaced Landau orbitals. The Coulomb interaction, however, correlates these states into the 2D magnetoexciton. The relative strength of the Coulomb and magnetic energies may be characterized by the dimensionless parameter $\lambda=(a_0/l_c)^2=\omega_c/2E_0$, where $a_0=\varepsilon_0/me^2$ and $E_0=me^4/2\varepsilon_0^2$ are the 3D exciton Bohr radius and Rydberg, and $l_c=(c/eH)^{1/2}$ and $\omega_c=eH/mc$ are the cyclotron radius and frequency, respectively. At high magnetic fields, $\lambda\gg1$, the importance of the Coulomb correlation relative to the magnetic-field effects is diminished.[2] In the limit $\lambda\rightarrow\infty$, when electrons and holes are confined to the lowest Landau orbital, 2D magnetoexcitons are exactly described by noninteracting point particles.[3] As for their nonlinear optical response, they should then behave like isolated two-level systems.[4]

The sample studied consists of 60 periods of 70-Å GaAs/AlGaAs QWs grown on GaAs. The growth substrate is removed by chemical etching to allow transmission experiments, and an antireflection coating applied to one surface to reduce Fabry-Pérot interference effects. For our nonlinear optical experiments, we use a synchronously pumped dye laser system that generates 100-fs optical pulses of microjoule energies at an 8-kHz repetition rate, with center wavelength at 805 nm.[5] The beam is focused into a jet of ethylene glycol to produce broadband nearly linearly polarized continuum pulses. The output from the continuum generator is split into pump and probe beams. Using a narrow-band interference filter, the pump excitation spectrum is selected. A variable-delay stage establishes the relative timing of the two pulses, as both are focused at nearly normal incidence onto the sample, located at the center of a superconducting magnet. The probe is then collected into an optical fiber and delivered to an optical multichannel analyzer for parallel detection. The entire magnet system, including the magnet itself and all optics for delivery of the pump and probe beams, is mounted as a unit to the same optical table as the laser system. All experiments are performed at 4 K.

The linear absorption spectrum versus magnetic field is shown in Fig. 1. At zero field, the 1*s* heavy-hole (hh) exciton lies at lower energy than the 1*s* light-hole (lh)

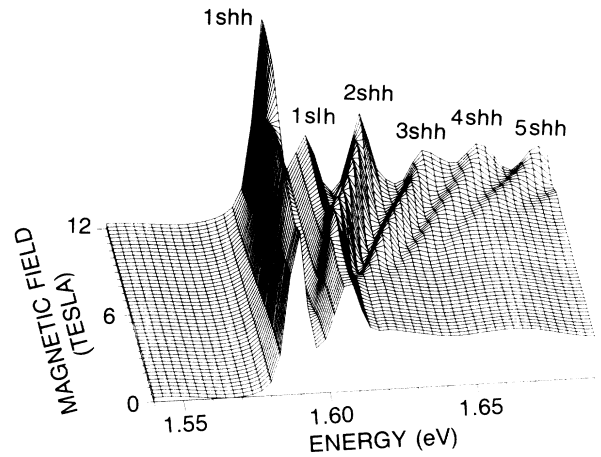

FIG. 1. Linear absorption data vs magnetic field. For clarity, one point is interpolated between each pair of data points along the magnetic-field axis.

exciton and the continua of electron-hole scattering states; all other bound states of the hh exciton lie within one linewidth of the 1s lh state, contributing to the absorption at that energy. The 1s hh and lh peaks disperse weakly with increasing magnetic field, showing only a small diamagnetic shift, while the 2s hh and higher excited states disperse more strongly, as expected[2] and seen earlier.[6] At fields around 6 T, the 2s hh peak moves above the 1s lh peak, after which point the ratio of oscillator strengths of 1s hh and lh transitions is close to the approximate theoretical value of 3. The excited-state transitions of the lh exciton are weaker than those of the hh by the same factor, and thus not resolved. As the magnetic field increases further, and l_c continues to decrease, the enhanced electron-hole overlap increases the oscillator strength of each resonance. The separation of the states increases simultaneously, conserving total oscillator strength. At 12 T, $\lambda \sim 3$ and $l_c \sim 70$ Å, and the lowest magnetoexciton states are strongly confined within the plane of the QW.

We investigate the consequences of the 0D confinement by measuring the transient nonlinear optical response of the magnetoexciton states as a real population is created in either the 1s hh state or the 2s hh state. The 0D response at $H = 12$ T is contrasted with the 2D response at $H = 0$ T. The optical Stark-effect response to a virtual population of 0D magnetoexcitons will be reported elsewhere.

In the first set of nonlinear optical experiments, with the sample at zero magnetic field, the pump beam was filtered so that only 1s hh excitons were created [Fig. 2(a)]. The resulting absorption spectrum [Fig. 2(b), inset] shows the 1s hh resonance when the probe arrives 600 fs before the pump (dashed) and 600 fs after (solid). Creation of a population of 1s hh excitons causes a loss of 1s hh oscillator strength, as one would expect due to phase-space filling.[1] The resonance is also shifted to the

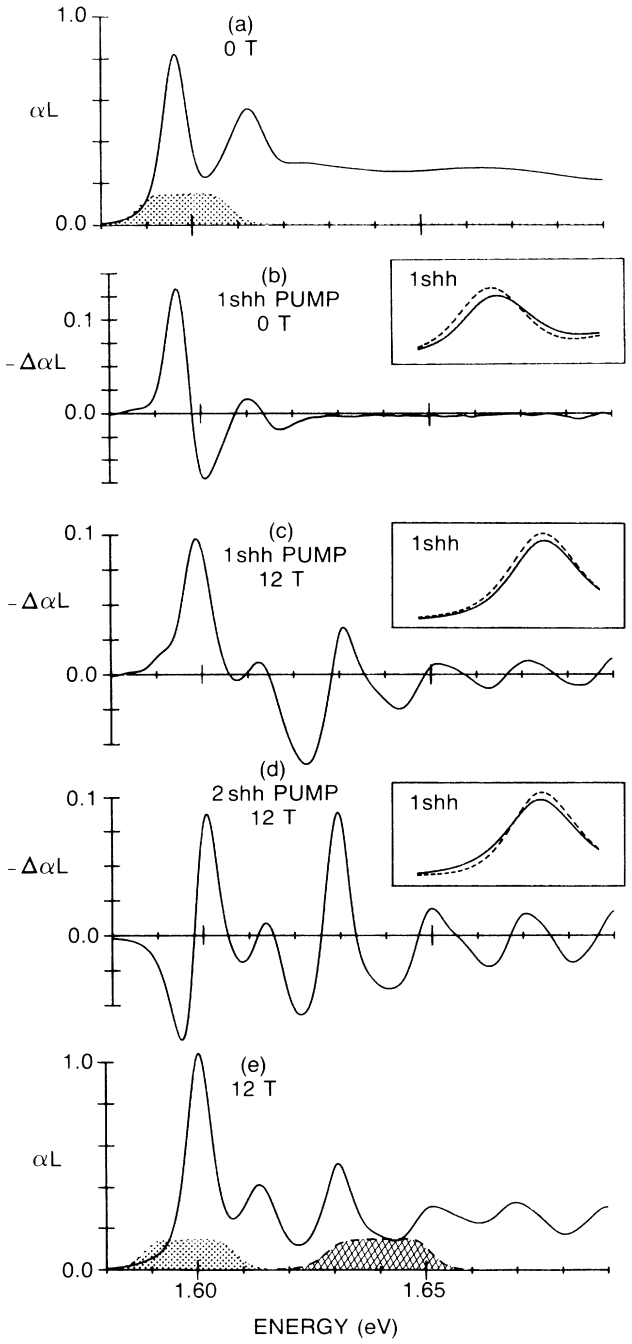

FIG. 2. Linear absorption at (a) 0 T and (e) 12 T. Dotted and hatched areas indicate the pump spectra for 1s and 2s hh exciton pumping, respectively. Differential transmission (b),(c) for 1s hh exciton pumping at 0 T and 12 T, respectively, and (d) for 2s hh exciton pumping at 12 T, all for probe 600 fs after pump. Insets: The corresponding 1s hh exciton absorption 600 fs before (dashed) and 600 fs after (solid) the arrival of the pump.

blue, corroborating earlier results for resonantly excited QWs.[7] This can be understood intuitively by considering the excitons to be hard spheres, which interact through a

steeply rising repulsive potential that acts to keep them from overlapping. When many such excitons are created by the pump, those excitons created by the probe experience the repulsive potential of all the others, which drives them to higher energy. The net result is a blueshift of the resonance.[1] In the differential transmission spectrum at zero field [Fig. 2(b)], the transmission on the low-energy side of the $1s$ hh resonance increases, while decreasing on the high-energy side, again indicating a blueshift of the resonance. Integrating the differential transmission spectrum across the $1s$ hh line demonstrates that there is a net loss of oscillator strength.

When a population of $1s$ hh magnetoexcitons is created at 12 T [Fig. 2(e), dotted area], the nonlinear optical absorption [Fig. 2(c), inset] also exhibits a loss of oscillator strength due to phase-space filling. The blueshift, however, is almost completely eliminated, indicating that the states have been confined into 0D and the intramagnetoexciton interaction quenched, as predicted theoretically.[3,4,8] With the $1s$ hh magnetoexciton interaction eliminated, the excitons behave as a gas of noninteracting particles. Additionally, the differential transmission at 12 T [Fig. 2(c)] shows nonzero signals over the entire spectral range. At the $2s$ hh resonance, the transmission decreases on the low-energy side of the resonance, while increasing on the high-energy side, indicating a redshift. At the higher excited magnetoexciton states, the differential transmission signals also derive from redshifts and collisional broadening due to intermagnetoexciton interaction with the $1s$ hh population.

In the second set of experiments, a population is created in the $2s$ hh magnetoexciton state [Fig. 2(e), hatched area]. The differential transmission spectrum versus time delay (Fig. 3) demonstrates coherent oscillations when the probe precedes the pump[9] and kinetic processes as the population created in the $2s$ hh state relaxes down into the $1s$ hh state. The effect on the $1s$ hh exciton of the population created in the $2s$ hh state is a redshift and bleaching [Fig. 2(d), inset]. The differential transmission signal at the $2s$ hh resonance is almost symmetric [Fig. 2(d)], indicating a loss of oscillator strength and collisional broadening of that state, with very little redshift. The higher-lying magnetoexciton states exhibit redshift and collisional broadening signals similar to those for $1s$ hh pumping. We note, however, that the broadening of the $1s$ hh exciton is much smaller than that of the higher excited states.

A qualitative understanding of the observed changes in the 12-T absorption spectrum can be obtained from the semiconductor density-matrix equations for interacting (purely) 2D electron-hole pairs in a perpendicular magnetic field and under the combined action of pump and probe pulses.[4] We have solved these equations numerically for various magnetic fields, time delays, and excitation conditions, and present results for 130-fs Gaussian pulses centered on the $1s$ and $2s$ magnetoexciton resonances. The probe pulse is linearly chirped, to make it broadband, and delayed by 520 fs. In order to simulate the effects of electron and hole form factors,[8] a slightly higher magnetic field, corresponding to $\lambda = 4$, was assumed.

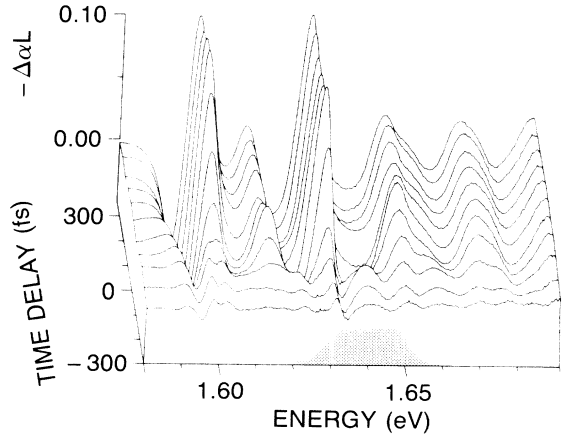

FIG. 3. Differential transmission data for $2s$ hh exciton pumping at 12 T vs time delay. Dotted area indicates pump spectrum.

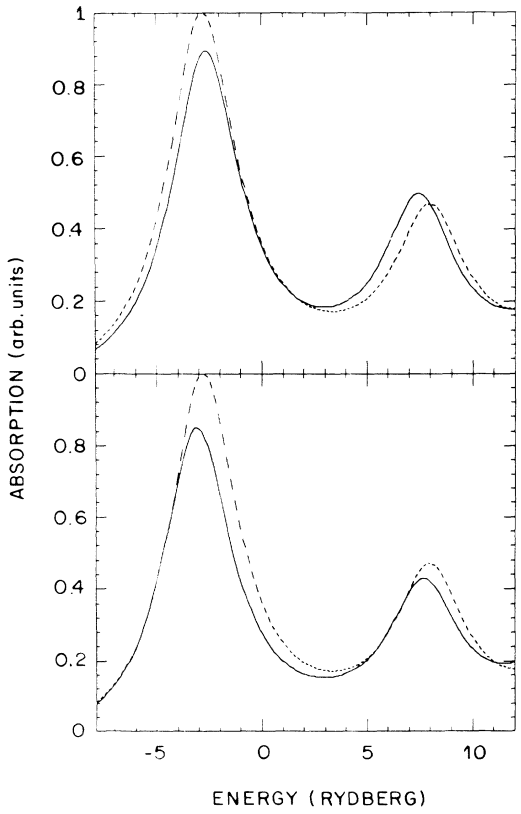

FIG. 4. Calculate (dashed) and nonlinear (solid) absorption for $1s$ (top) and $2s$ exciton (bottom) pumping.

Figure 4 shows the calculated changes in the absorption spectrum for $1s$ (top) and $2s$ (bottom) magnetoexciton pumping with peak Rabi frequencies of $0.2E_0$ and $0.4E_0$, respectively. Although dephasing processes are treated phenomenologically ($T_2^{-1}=2E_0$), which cannot describe the observed collisional broadening, the theoretical results agree rather well with the experimental ones. For $1s$ excitation, the saturation of the $1s$ exciton resonance is accompanied by a very small blueshift. We find that this blueshift decreases with increasing magnetic field. The shift of the $2s$ exciton state, however, is to lower energies; it corresponds to the so-called band-gap renormalization in zero field,[1] since at high magnetic fields, the relative change of the excited-magnetoexciton-state wave functions, due to the electron-hole Coulomb attraction, is small.[2,10] Conversely, $2s$ excitation causes, in addition to saturation, a redshift of the $1s$ exciton resonance, showing that the intermagnetoexciton interaction is attractive, in agreement with experiment. The stronger collisional broadening of the higher magnetoexciton states, relative to that of the $1s$ hh state, may also be understood by considering the fact that $1s$-$1s$ scattering has a smaller collision cross section than that of $1s$-ns ($n=2,3,\ldots$), because of the smaller spatial extent of the $1s$ exciton wave function.

In summary, we report the first measurement of the femtosecond-time-resolved nonlinear optical response of magnetically confined quantum-well states, as their dimensionality is continuously tuned from 2D to 0D. At low fields, a population created in the $1s$ hh magnetoexciton state causes a blueshift and bleaching of that state due to intramagnetoexciton interaction. At high fields, when the system is 0D, creation of a population in the $1s$ hh or $2s$ hh state causes a bleaching of that state, with almost no energy shift, a redshift of unpopulated states, and collisional broadening. The observed nonlinear optical response agrees qualitatively with calculations describing Coulomb-coupled Landau orbitals.[4] Most importantly, these measurements demonstrate for the first time the remarkable and exact theoretical result that at high magnetic fields an ensemble of $1s$ magnetoexcitons behaves like a gas of noninteracting particles.[3] This behavior is unique among many-body systems.

We acknowledge the expert assistance of J. E. Henry in the preparation of the quantum-well sample, and C. W. Tu for its growth. We would also like to thank Peter. A. Wolff for many helpful discussions. One of us (J.B.S.) would like to acknowledge support through NSF Grant No. EET-871841.

[a]Permanent address: Physics Department, Massachusetts Institute of Technology, Cambridge, MA 02139.

[b]Permanent address: Hochstleistungsrechenzentrum, Forschungszentrum Jülich, D-5170 Jülich, Federal Republic of Germany.

[c]Permanent address: Physics Department, Princeton University, Princeton, NJ 08544.

[1]S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Adv. Phys. **38**, 89 (1989).

[2]O. Akimoto and H. Hasegawa, J. Phys. Soc. Jpn. **22**, 181 (1967); M. Shinada and K. Tanaka, J. Phys. Soc. Jpn. **29**, 1258 (1970); A. H. MacDonald and D. S. Richie, Phys. Rev. B **33**, 8336 (1986); S. R. E. Yang and L. J. Sham, Phys. Rev. Lett. **58**, 2598 (1987).

[3]I. V. Lerner and Yu. E. Lozovik, Zh. Eksp. Teor. Fiz. **80**, 1488 (1981) [Sov. Phys. JETP **53**, 763 (1981)]; D. Paquet, T. M. Rice, and K. Ueda, Phys. Rev. B **32**, 5208 (1985).

[4]C. Stafford, S. Schmitt-Rink, and W. Schäfer, Phys. Rev. B **41**, 10000 (1990).

[5]W. H. Knox, J. Opt. Soc. Am. B **4**, 1771 (1987).

[6]S. Tarucha, H. Okamoto, Y. Iwasa, and N. Miura, Solid State Commun. **52**, 815 (1984).

[7]N. Peyghambarian, H. M. Gibbs, J. L. Jewell, A. Antonetti, A. Migus, D. Hulin, and A. Mysyrowicz, Phys. Rev. Lett. **53**, 2433 (1984); W. H. Knox, R. L. Fork, M. C. Downer, D. A. B. Miller, D. S. Chemla, C. V. Shank, A. C. Gossard, and W. Wiegmann, Phys. Rev. Lett. **54**, 1306 (1985); S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Phys. Rev. B **32**, 6601 (1985).

[8]G. E. W. Bauer, Phys. Rev. Lett. **64**, 60 (1990).

[9]C. H. Brito-Cruz, J. P. Gordon, P. C. Becker, R. L. Fork, and C. V. Shank, IEEE J. Quantum Electron. **24**, 261 (1988); B. Fluegel, N. Peyghambarian, G. Olbright, M. Lindberg, S. W. Koch, M. Joffre, D. Hulin, A. Migus, and A. Antonetti, Phys. Rev. Lett. **59**, 2588 (1987).

[10]For $1s$ excitation in zero field, the $2s$ exciton experiences a small blueshift as discussed by G. W. Fehrenbach, W. Schäfer, and R. G. Ulbrich, J. Lumin. **30**, 154 (1985).

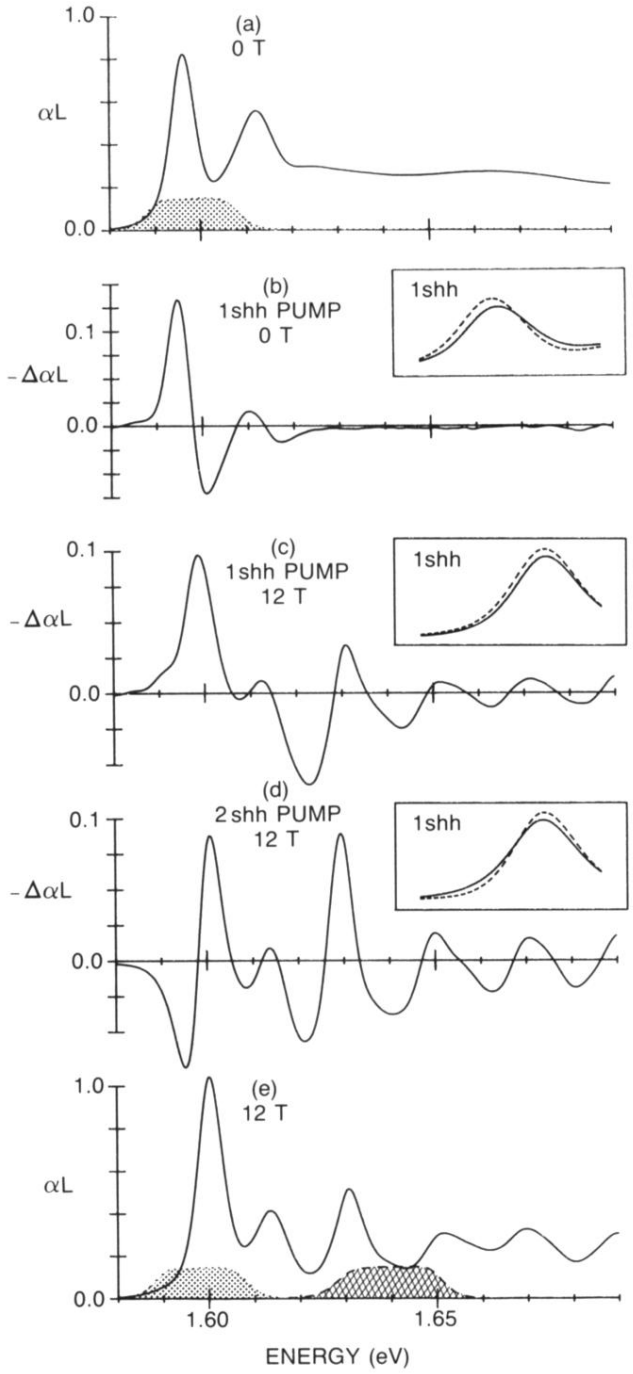

FIG. 2. Linear absorption at (a) 0 T and (e) 12 T. Dotted and hatched areas indicate the pump spectra for 1*s* and 2*s* hh exciton pumping, respectively. Differential transmission (b),(c) for 1*s* hh exciton pumping at 0 T and 12 T, respectively, and (d) for 2*s* hh exciton pumping at 12 T, all for probe 600 fs after pump. Insets: The corresponding 1*s* hh exciton absorption 600 fs before (dashed) and 600 fs after (solid) the arrival of the pump.

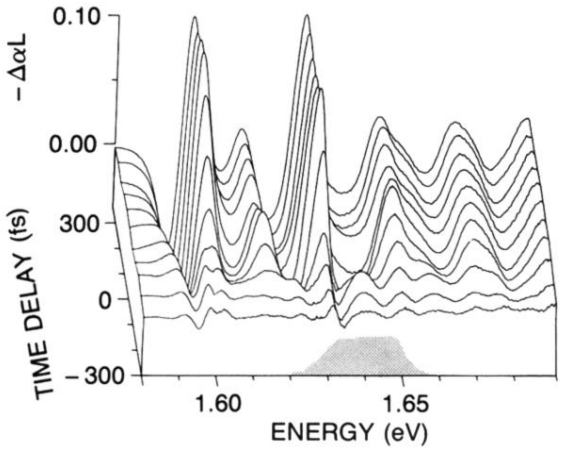

FIG. 3. Differential transmission data for $2s$ hh exciton pumping at 12 T vs time delay. Dotted area indicates pump spectrum.