cw and Femtosecond Optical Nonlinearities of Type-II Quantum Wells

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Femtosecond time-resolved and quasi-cw measurements of optical nonlinearities of type-II GaAs/AlAs quantum wells are reported. A pronounced blueshift of the heavy-hole exciton is observed, which develops on a 100-ps time scale after resonant interband excitation. The experimental observations are analyzed using many-body theory, explaining exciton blueshift and dynamical evolution in terms of electron-hole Coulomb enhancement, hole-phase-space filling, and hole-plasma cooling.

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The influence of an electron-hole (e-h) plasma on the optical absorption of highly excited semiconductors has been studied extensively both experimentally and theoretically in bulk semiconductors and type-I quantum-well systems.[1-5] When Feldmann *et al.*[6] reported on the spatial separation of electrons and holes in a type-II GaAs/AlAs quantum-well structure, it became apparent that these systems allow the investigation of plasma nonlinearities for the case where only a single-component plasma affects the absorption. Since e-h charge separation occurs within less than a ps, optical excitation of a type-II structure leads to formation of a one-component plasma on a sub-ps time scale, the dynamics of which can then be observed optically on a ps to ns scale.

In this Letter we report measurements of cw and fs nonlinearities of type-II systems. We analyze the results in terms of a many-body theory similar to the well-known e-h plasma theory of bulk semiconductors.[1-5] We show that the absence of one plasma component changes drastically the optical nonlinear behavior of a semiconductor layer. One direct consequence of the absence of one plasma component is the absence of optical gain as there are no electrons available for stimulated emission. Furthermore, a blueshift of the heavy-hole (HH) exciton is observed and identified as the so-called Mahan exciton, i.e., the Coulomb enhancement peak at the quasi-chemical-potential energy.

We investigated three different type-II structures labeled as (11/30), (11/18), and (11/11), respectively, where (n/m) denotes n monolayers of GaAs and m monolayers of AlAs, and one monolayer equals 2.83 Å. The Γ-X splittings of our three structures are 95, 80, and 61 meV, respectively, obtained as the energy difference between Γ-Γ and Γ-X peaks in photoluminescence measurements.

We first present the quasi-cw nonlinear absorption spectra for the (11/30) and (11/11) structures in Fig. 1. Here we used modulated-pump and probe spectroscopy to collect excitation-intensity-dependent quasi-steady-state absorption spectra, where time-correlated continuum probe pulses were delayed 10 ns with respect to pump pulses. The spectra in Fig. 1 show that the HH exciton resonance shifts significantly toward higher energies and bleaches with increasing excitation intensity. In contrast, the light-hole (LH) exciton shifts very slightly to lower energies and exhibits little reduction in oscilla-

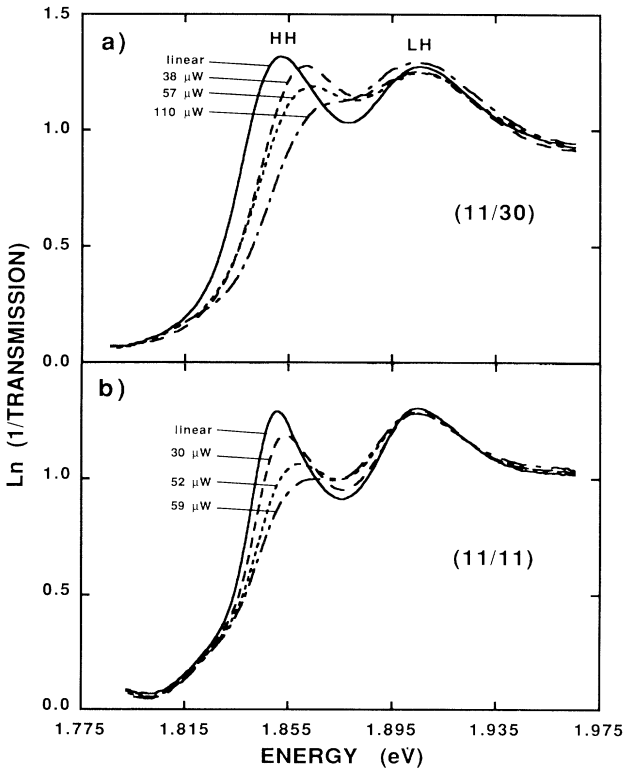

FIG. 1. Excitation-intensity-dependent absorption spectra of structure (a) (11/30) and (b) (11/11). $T=15$ K, $E_{\text{laser}}=2.14$ eV, $f=8.2$ kHz, spot size $=100$ μm, and $t_{\text{pump}}=10$ ns. The carrier density for the spectrum labeled 110 μW is $\simeq 10^{12}/\text{cm}^2$.

tor strength. Comparing the amount of blueshift in the (11/30) and (11/11) samples shown in Fig. 1 and in the (11/18) sample (not shown), we observe a decrease and eventual saturation of the intensity-dependent blueshift of the HH exciton with decreasing Γ-X splitting.

In Fig. 2 we present fs pump-probe spectra of the (11/30) structure obtained using 100-fs pump pulses and a time-correlated white-light continuum. We observe a small blueshift ($\simeq$4 meV) of the HH exciton for pump-probe time delays after several hundred femtoseconds which then develops into the large blueshift (>15 meV) as the quasiequilibrium state is approached [notice the strong similarity between the 100-ps curve in Fig. 2 and the 110-μW curve in Fig. 1(a)].

The observed nonlinearities are caused by the excited-hole and -electron plasmas which separate on a sub-ps time scale with respect to both spatial and momentum coordinates.[6-8] The cooling of the plasmas occurs on a $\sim$100-ps time scale, so that the quasi-steady-state condition is that of a cold e plasma located at the X point of the Brillouin zone in the AlAs layer and a cold HH plasma at the Γ point in the GaAs layer.

In bulk structures an increasing e-h density first leads to a pure bleaching of the exciton without any significant shift until, at high densities, a gain region occurs. The crossover point from gain to absorption (i.e., the quasi-chemical-potential of the e-h plasma) shifts to higher energies with increasing density since the band-gap shrinkage due to the plasma is always less than the increase of the chemical potential. The absorption spectrum around the chemical potential experiences significant Coulomb enhancement. This peak is the so-called Mahan exciton, which has been discussed for type-I and n-doped semiconductor systems.[3] In type-I structures the simultaneous occurrence of optical gain clearly distinguishes the Mahan exciton peak from the low-density exciton (bound e-h pair) resonance. Because of the absence of gain in a type-II system the Mahan exciton resonance above the quasi-chemical-potential appears as a blueshifted exciton resonance. We verified the absence of gain in type-II samples by comparing the nonlinear absorption spectra of the (11/30) type-II structure with a type-I quantum-well structure consisting of 24-Å-thick GaAs wells and 100-Å-thick $Al_{0.45}Ga_{0.55}As$ barriers. Using the same optical excitation conditions for both samples we observed gain ($\alpha_{max}=-0.2$, $\Delta E_{\text{gain band}}\simeq 50$ meV) in the type-I structure and a corresponding zero-absorption region in the type-II structure.

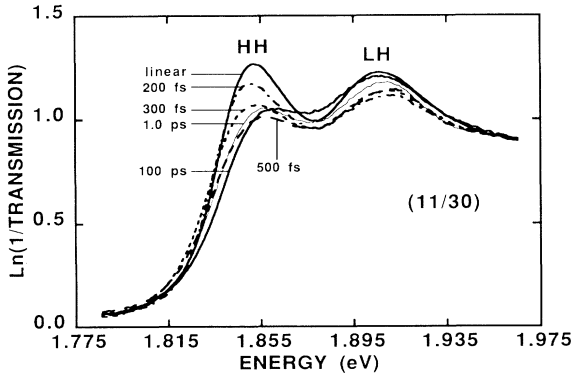

FIG. 2. Femtosecond time-resolved absorption spectra for the (11/30) structure. $P_{exc}=8.5$ μW (corresponding to a carrier density of $\sim 1\times 10^{12}/cm^2$), $E_{exc}=1.96$ eV, and $T=15$ K. The uncertainty in the peak intensity for Figs. 1 and 2 is $<40\%$.

In order to compute the discussed nonlinearities, we proceed in two steps. First, we calculate the shifts of the in-plane valence and conduction bands caused by the induced space-charge field by solving the Poisson and Schrödinger equations for the motion perpendicular to the layers. A strong voltage drop between neighboring layers develops[8] for carrier densities $\sim 10^{12}/cm^2$, but the space-charge field has little effect on the valence-conduction-band separation within the same layer since both bands shift essentially parallel. The oscillator strength of the HH transition changes by less than 1%. Note that this insensitivity of the absorption to electric-field effects differs from the case of n-i-p-i structures.[9] Even for the highest densities investigated the induced electric fields in the type-II structures cause band-gap shifts of less than 2 meV to higher energies, hence, leading only to small modifications of the optical absorption. In contrast, the space-charge-field modifications of the luminescence are large and have been reported in Ref. 8.

In the second step of our calculations we compute the absorption spectra of the GaAs layer, assuming perfect e-h charge separation. The LH band is unoccupied for our densities so that only the HH occupation contributes to phase-space filling. These model assumptions are best justified for the (11/30) structure since at $T=15$ K and the densities under consideration the difference between the chemical potentials of the electrons in the AlAs layer and the Γ point in the GaAs layer is several k_BT. Neglecting the HH-LH exchange coupling, the equations for the interband polarization P_k for LH and HH transitions are decoupled:

$$\left\{\epsilon_k^c-\epsilon_k^v+\sum_{k'}\Delta W_{k'k}-\hbar\omega-i\gamma\right\}P_k - \sum_{k'}[V_{k-k'}+\Delta W_{kk'}]P_{k'}=M_k^v f_k^v\,, \tag{1}$$

where ϵ_k^c and ϵ_k^v are conduction- and valence-band energies with v denoting either the HH or the LH valence band, ω is the probe frequency, k is the in-plane momentum, γ accounts for linewidth broadening, M_k^v is the optical matrix element, f_k^v is the Fermi distribution function of the valence-band electrons, and $V_{kk'}$ is the bare Coulomb potential of a quantum well with infinite bar-

rier height accounting for the lowest subband only. Exchange effects due to the Pauli principle and screening due to the HH plasma are described by

$$\Delta W_{kk'} = V_{k-k'}(f_k^v - 1 + \chi_{kk'}^v - \chi_{kk'}^c), \quad (2)$$

with the dynamical screening function

$$\chi_{kk'}^i = \frac{\omega_{pl}^2(q)}{2\omega_q}\left[\frac{f_k^i + g(\omega_q)}{E_k^i - \Delta_{k'} - \omega_q} - (\omega_q \leftrightarrow -\omega_q)\right], \quad (3)$$

where $i = c,v$ and $\mathbf{q} = \mathbf{k} - \mathbf{k}'$, $E_k^i = \epsilon_k^i \pm (E_g + i\gamma)/2$, $(+,-$ for $v,c)$, $g(\omega_q)$ is the plasmon Bose distribution function, and $\Delta_k = (\epsilon_k^c + \epsilon_k^v)/2$. The function χ describes the absorption and emission of quasi-two-dimensional plasmons of the HH plasma with plasma frequency $\omega_{pl}(q)$ and dispersion ω_q. Equation (3) is an extension of the screened Hartree-Fock quasiparticle approximation of the single-particle self-energy to the case of excitons, which avoids the artificial singularities associated with the so-called Shindo approximation[10] which become particularly questionable for type-II systems. Extending the numerical methods of Ref. 11 we solve Eqs. (1)–(3) to obtain the homogeneous band-edge spectra for the HH and LH transitions. The total homogeneous absorption is obtained by summing the HH and LH contributions.

As may be seen from Fig. 1, the experimentally obtained resonances are relatively broad, indicating significant inhomogeneous broadening due to well-width fluctuations. We model these fluctuations by averaging the spectra using a Gaussian distribution of well-width thicknesses. The amount of broadening needed to reproduce the experimental zero-density spectrum corresponds to well-width fluctuations of about half a monolayer.[12] Figure 3(a) shows our theoretically computed carrier-density-dependent spectra exhibiting good qualitative agreements with those of Fig. 1(a). The corresponding spectra without inhomogeneous broadening [see Fig. 3(b)] reveal that the observed HH exciton blueshift is in fact the transition to the Mahan exciton at the Fermi edge. For all densities shown, the reduced HH band gap is below the zero-density HH-exciton resonance. For the LH exciton we find a redshift with relatively little loss in oscillator strength. This shift is caused by the screening due to the HH plasma (generalized Coulomb hole).

We explain the delayed onset and full development of the magnitude of the blueshift in the femtosecond measurements of Fig. 2 through thermalization of the electrons and holes. Initially electrons and holes are injected into the conduction and valence bands with considerable excess energy. After the initial rapid cooling by LO-phonon emission, complete thermalization to the lattice temperature occurs on the time scale of hundreds of pi-

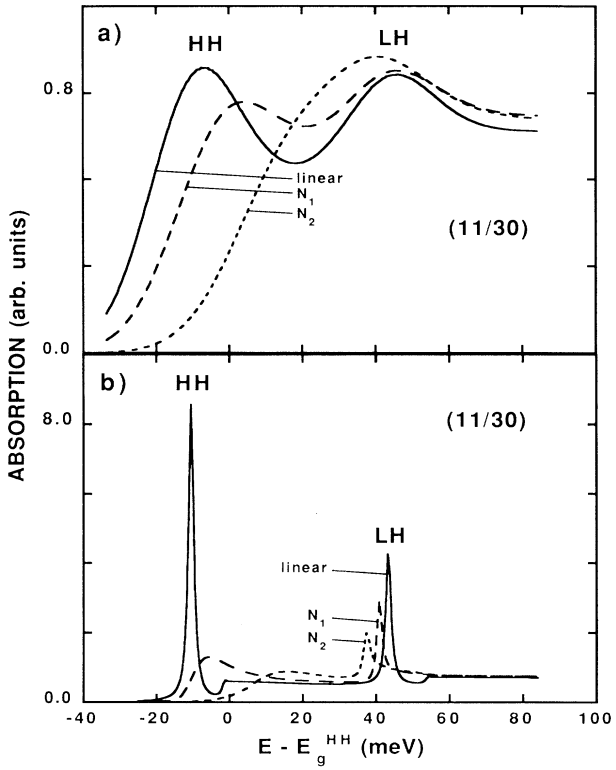

FIG. 3. (a) Calculated carrier-density-dependent absorption spectra computed for a (11/30) GaAs type-II quantum well including inhomogeneous broadening through well-width fluctuations. Linear ($N \simeq 0$) spectrum, and spectra for the carrier densities $N_1 = N_0$ and $N_2 = 3.0N_0$, where $N_0 = 0.3a_B^{-2}$, a_B is the bulk GaAs Bohr radius. (b) Corresponding linear and $N = N_0$ spectra without inhomogeneous broadening. E_g^{HH} is the HH energy gap and $T = 15$ K.

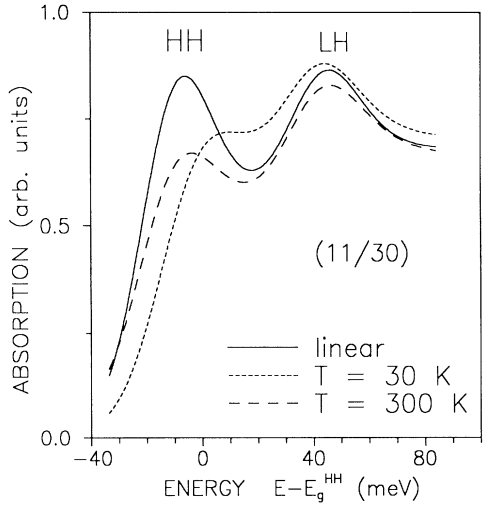

FIG. 4. Calculated carrier-density-dependent absorption spectra computed for a (11/30) GaAs type-II quantum well including inhomogeneous broadening through well-width fluctuations. Linear ($N \simeq 0$) spectrum and spectra for the carrier density $N_0 = 0.4a_B^{-2}$ for hole-plasma temperatures $T = 30$ and 300 K.

coseconds via acoustic-phonon emission. To verify this conjecture we show in Fig. 4 computed inhomogeneously broadened spectra for a fixed hole-plasma density at 30 and 300 K. Comparison of the 300-K curve with the 300-fs spectrum in Fig. 2 and of the 30-K curve with the 100-ps spectrum in Fig. 2 yields good qualitative agreement. Hence, we see that for a hot plasma there is little blueshift of the exciton resonance due to phase-space filling since the carriers have not yet relaxed to the band minimum. This feature has been verified experimentally by ns experiments at liquid-nitrogen temperatures showing the disappearance of the blueshift.[13]

In conclusion, we observed and analyzed femtosecond-time-resolved and quasi-cw optical nonlinearities in type-II quantum wells. As unique consequences of the *e-h* charge separation in these structures we observe the absence of optical gain and modified optical nonlinearities, such as a blueshift of the heavy-hole exciton, which are caused predominantly by the hole plasma in the GaAs layers. We also observed a delayed onset of the full development of the HH exciton blueshift due to slow cooling of the HH plasma. It is worthwhile to note that a blueshift of the exciton absorption in narrow type-I quantum wells has been observed[14] and explained in terms of the existence of an exciton gas rather than an *e-h* plasma.[14,15] Clearly, this explanation cannot be used for our type-II systems since the *e-h* charge separation inhibits formation of an exciton gas in the GaAs layer.

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