Laser-Induced Exciton Splitting

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A laser beam irradiating a semiconductor in its transparency region induces a splitting of the exciton line through the dependence of the optical Stark effect on the different transition matrix elements. The splitting is observed in bulk and multiple-quantum-well GaAs, by femtosecond time-resolved spectroscopy for various polarization configurations. Experimental results are in good agreement with theory.

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The interaction of semiconductors with below-gap laser beams[1,2] has recently become of large interest[3-10] and has shed new light upon virtual excitations. Development of ultrashort, intense laser pulses allows us to now extend, to solid-state physics, effects first reported in atomic physics[11] and bring up fruitful results due to the complexity of optical transitions in semiconductors. In this Letter, we claim that laser beams not only shift but also split the exciton level. We give the theoretical origin of this splitting and we report its first experimental observation. We also explain why the optical Stark shift does not depend on the linear beam polarization in multiple-quantum-well structures (MQWS).[1,4]

The optical Stark effect shows up mainly as a shift towards higher energy of the absorption lines. This shift is inversely proportional to the detuning, i.e., the energy difference between absorption line and photon. It is also proportional to the square of the optical matrix element. This last feature is at the origin of the light-induced splitting of a degenerate exciton: As shift amplitudes differ for the various optical transitions, it allows their separation. Splittings are usually obtained by uniaxial stress, magnetic or electric fields, but this is here, to our knowledge, the first case where light is used to provide such an effect.

Combescot and Combescot[9] have recently shown that the exciton shifts can be calculated without the Coulomb interaction when the detuning is large compared with the exciton binding energy. In these conditions the detuning is almost the same for all transitions (towards bound and unbound electron-hole states) and one expects rigid valence- and conduction-band shifts. This remark is quite useful as it avoids many-body treatments and allows us to consider only transitions from one free valence state to one free conduction state. The situation is then similar to a two-level system in atomic physics, except that we have now a multilevel atom. The conduction band being spin degenerate and the valence band resulting from a p-type spin-orbit coupling, one expects to find at large detuning the shifts of a $(2+2\times 3)$ i.e., eight-level, dressed atom.[10] Let us now consider this problem.

The electron-photon interaction reads $W^\dagger + W$, where $W^\dagger$ is a spin-conserving operator, $W^\dagger = \sum_s W_s^\dagger$;

$$W_s^\dagger = (qP/m) a_s^\dagger [A_x b_{x,s} + A_y b_{y,s} + A_z b_{z,s}] , \qquad (1)$$

where $s = \pm\frac{1}{2}$, x, y, and z are the sample cubic axes, P the Kane matrix element, and A_x the field-potential component along x. $a_s^\dagger$ creates a conduction electron with spin s. $b_{x,s}$ destroys a valence electron with "x" symmetry and s spin. For simplicity, we will consider only light propagating along the z axis, i.e., $A_z = 0$. If we now take into account the spin-orbit coupling, the valence operators are preferably rewritten in terms of b_m $(m = \pm\frac{3}{2}, \pm\frac{1}{2})$ for the $j = \frac{3}{2}$ states and b'_m $(m = \pm\frac{1}{2})$ for the $j = \frac{1}{2}$ states. Using this new basis, the electron-photon coupling is the following:

$$W^\dagger_{\pm 1/2} = a^\dagger_{\pm 1/2} \left[\mp \lambda_\mp b_{\pm 3/2} \pm \sqrt{\tfrac{1}{3}}\, \lambda_\pm b_{\mp 1/2} - \sqrt{\tfrac{2}{3}}\, \lambda_\pm b'_{\mp 1/2} \right] , \qquad (2)$$

with $\lambda_\pm = qP(A_x \mp iA_y)/m\sqrt{2}$. As expected, Eq. (2) just tells us that, for example, the σ_+ photon (corresponding to λ_+) induces transitions from valence to conduction states with $\Delta m = +1$ (namely from $-\frac{3}{2}$ to $-\frac{1}{2}$ and from $-\frac{1}{2}$ to $+\frac{1}{2}$).

The shifts of the $(2+6)$ electron states induced by the pump beam are easily calculated from second-order perturbation theory. The valence-conduction coupling pushes up the two conduction states $a_{\pm 1/2}$ by an amount $\Delta_\pm$:

$$\Delta_\pm = \lambda^2_\mp/\Omega_H + \lambda^2_\pm/3\Omega_L + 2\lambda^2_\pm/3\Omega' = \delta_{H\pm} + \delta_{L\pm} + \delta'_\pm , \qquad (3)$$

TABLE I. Shifts Δ_i (in units of λ^2/Ω_H) and relative weights η_i of the exciton line for bulk GaAs and MQWS (for the heavy- and light-hole exciton) with pump and probe beams parallel to the z axis with varying polarizations $\rho = \Omega_H/\Omega_L$.

	Pump	Probe	GaAs bulk				MQWS (H)		MQWS (L)	
Δ_i	$\sigma+$		$\frac{3}{2}$	$\frac{1}{2}$	3	1	2	$\rho/3$	$2\rho/3$	1
η_i		$\sigma+$	0	0	$\frac{3}{4}$	$\frac{1}{4}$	1	0	1	0
		$\sigma-$	$\frac{1}{4}$	$\frac{3}{4}$	0	0	0	1	0	1
		linear	$\frac{1}{8}$	$\frac{3}{8}$	$\frac{3}{8}$	$\frac{1}{8}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$
Δ_i	linear		1	2			$2+\rho/3$		$1+2\rho/3$	
η_i		parallel	0	1			1		1	
		perpendicular	$\frac{3}{4}$	$\frac{1}{4}$			1		1	
		$\sigma\pm$	$\frac{3}{8}$	$\frac{5}{8}$			1		1	

Ω_H, Ω_L, and Ω' being the detunings with respect to the various valence states. H and L stand for heavy and light holes ($m = \pm\frac{3}{2}, m = \pm\frac{1}{2}$). Similarly, the valence-conduction coupling pushes down the valence states. Their shifts depend on whether these states are degenerate or not: (i) If $\Omega_H < \Omega_L < \Omega'$ (as for MQWS), the states $b_{\pm 3/2}$, $b_{\mp 1/2}$, and $b'_{\mp 1/2}$ are respectively shifted by an amount $-\delta_{H\pm}$, $-\delta_{L\pm}$, and $-\delta'_{\pm}$. (ii) If $\Omega_H = \Omega_L < \Omega'$ (as for bulk GaAs), the four states $b_{\pm 3/2}$ and $b_{\pm 1/2}$ are degenerate. Two linear combinations of these states are coupled by the light to the conduction states, while the two orthogonal ones are not [see Eq. (2)], their shifts being $-(\delta_{H\pm} + \delta_{L\pm})$ if the detuning is small compared with the spin-orbit splitting (the b'_m then do not play any role).

The excitonic shifts induced by the pump beam at large detuning[9] are simply the difference between conduction and valence state shifts. (i) For GaAs quantum wells ($\Omega_H < \Omega_L \ll \Omega'$), the heavy- (respectively, light-) hole fourfold exciton splits into four distinct levels, which are blue shifted by $2\delta_{H\pm} + \delta_{L\pm}$ and $\delta_{H+} + \delta_{H-} + \delta_{L\pm}$ (respectively, $2\delta_{L\pm} + \delta_{H\pm}$ and $\delta_{L+} + \delta_{L-} + \delta_{H\pm}$). (ii) For bulk GaAs ($\Omega_H = \Omega_L \ll \Omega'$) the eightfold exciton splits into five distinct levels which are blue shifted by $2\Delta_+$, $2\Delta_-$, Δ_+, Δ_-, and $\Delta_+ + \Delta_-$. The crossover region between MQWS and bulk behavior corresponds to shifts of the order of the heavy-light hole splitting.

Pump and probe beams with appropriate polarizations are used to see these various shifted exciton levels. The electron-probe photon coupling is similar to Eq. (2), with λ replaced by λ' related to the probe components. The weights η_i of the various shifted lines are easily obtained from the Fermi "golden rule." Predicted values are listed in Table I.

The experiments have been performed in bulk GaAs ($\Omega_H = \Omega_L$) and in GaAs-GaAlAs MQWS ($\Omega_H < \Omega_L$). Short tunable pulses[12] have been used to get sufficiently high intensity. The pump beam is an amplified part of the wavelength continuum generated in water by 60-fs pulses, leading to intensities up to 10^{10} W/cm^2 in the near-infrared region. More precisely, the pump is selected in the continuum with interferential filters of typically 50-Å bandwidth and further amplified in a styryl-9 dye cell. The chosen wavelengths were here respectively 827 and 853 nm for the MQWS and bulk experiments. Pump and probe beams are collinear with a propagation axis along the growth axis for both samples. The pump is polarized either linearly or circularly while the probe polarization is analyzed after the sample. The intensity of the pump is kept as low as possible to stay in the small-signal (perturbative) regime.

The optical Stark effect splits the exciton but the small magnitude of the shifts, with respect to the exciton linewidth, prevents a direct observation of the splitting. Fortunately, the various exciton levels are not reached with the same weights by optical transitions. A precise analysis of the shifts is performed with differential transmittance spectra. This sensitive method measures the difference between the unperturbed absorption line and the pump-perturbed one:

$$-\Delta\alpha(\omega) = \alpha(\omega) - \sum_i \eta_i \alpha(\omega - \Delta_i) \approx \left[\sum \eta_i \Delta_i\right] d\alpha/d\omega . \quad (4)$$

The shifts Δ_i are those induced by the pump, the relative weights η_i depend on both the pump and the probe (see Table I), and $d\alpha/d\omega$ is extracted from the bare experimental absorption spectrum $\alpha(\omega)$. The use of various pump and probe polarizations allows us to recover the original splittings hidden inside the exciton linewidth and to check the validity of the theory. Note that the observation of a signal change with probe polarization, while keeping identical pump parameters, is already a proof of the exciton splitting.

In MQWS, the optical Stark effect does not depend on the respective linear pump and probe polarizations as already reported,[1] in agreement with our theory (see Table I).[13] On the other hand, there is a strong dependence for

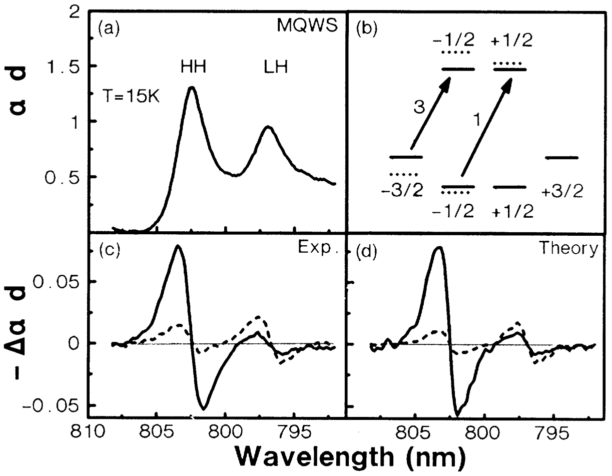

FIG. 1. (a) Absorption spectrum of a GaAs/AlGaAs MQWS sample with 50 wells and barriers each of thickness 100 Å. (b) Couplings induced by a σ_+ pump. The labels indicate the components of the kinetic momentum along the growth axis. The σ_+ pump induces selective couplings with relative strengths 3 and 1. The unperturbed levels are shown by solid lines, the shifted ones by dotted lines. (c) Differential transmittance spectra as measured by a σ_+ (solid line) and σ_- (dashed line) polarized probe. The pump beam has a σ_+ polarization and is tuned at 827 nm with a spectral width of 5.2 nm. The pump intensity is of the order of 100 MW/cm^2, several orders of magnitude larger than the probe. (d) Theoretical spectra for σ_+ (solid line) and σ_- (dashed line) probe polarization obtained from the derivative of the absorption spectrum (a) multiplied by the coefficients of Table I. The vertical scale is arbitrary.

circular σ_+ pump and σ_+ or σ_- probes (see Fig. 1). The small signal amplitude (less than 10%, corresponding to a shift of 0.2 meV) insures the validity of the perturbative regime for which the theory has been developed. Figure 1(b) indicates the shifts of the two transitions induced by a σ_+ pump, and their relative strengths. A σ_+ probe couples valence and conduction states differing by $\Delta m = +1$, both shifted by the σ_+ pump. On the other hand, a σ_- probe can only see the shifts of the conduction band as it induces only transitions with $\Delta m = -1$. Theoretical spectra are in very good agreement with experiments, although heavy-light hole band mixing[14] is not included.

In bulk GaAs, unlike in quantum wells, the shifts depend on the linear polarizations (Fig. 2). We measure a parallel-to-perpendicular signal ratio varying from 1.3 to 1.5 with increasing pump intensity (Table I gives $\frac{8}{5}$). For circular polarizations, the σ_+ to σ_- ratio is found to range from 2 in the low regime to 2.5 at higher regimes (Table I gives $\frac{10}{3}$). The discrepancy at small shift can be due to a splitting already present (for instance induced by strain), as the theory predicts a bulk behavior only for a Stark shift larger than the valence splitting.

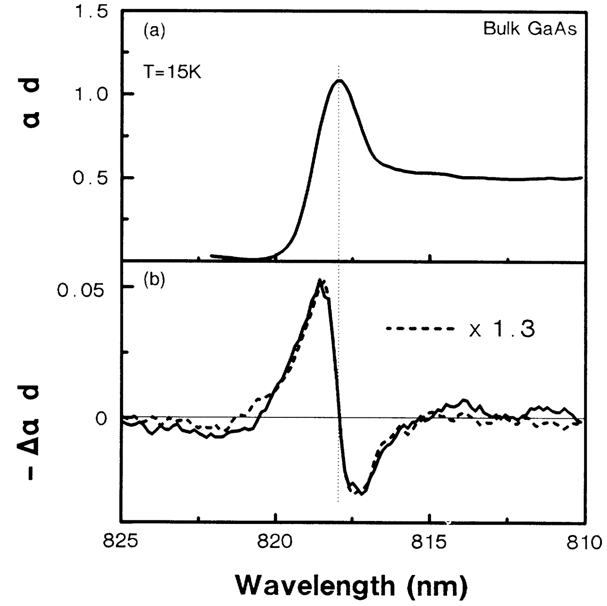

FIG. 2. (a) Absorption spectrum of the 0.5-μm-thick GaAs sample at 15 K. (b) Differential transmittance spectra for linearly polarized pump and probe either parallel (solid line) or perpendicular (dashed line). The dashed curve has been multiplied by 1.3 (see text). The pump at 853 nm has a spectral width of 6.5 nm.

Agreement between theory and experiments is good in MQWS. For bulk GaAs, it is better for a moderate light intensity than for a very low one. This suggests that this method could be a sensitive tool to reveal yet undetectable splitting of an absorption line. In the case of multiple quantum wells, this could also lead to the determination of the in-plane band nonparabolicity.

In conclusion, we show for the first time that a laser beam in the transparency region induces exciton splittings. These splittings, hidden inside the exciton linewidth, are observed in GaAs bulk and MQWS with time-resolved differential transmittance spectra for different pump and probe polarizations. The observed shifts are in quantitative agreement with the "dressed multilevel atom" approach. These results also provide the first experimental support of the new theory on the exciton optical Stark shift based on a dressed-atom picture at large detuning.[9]

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