OPTICAL STARK EFFECT IN GaAs QUANTUM WELLS

A. Mysyrowicz, D. Hulin

OPTICAL STARK EFFECT IN GaAs QUANTUM WELLS

A. MYSYROWICZ and D. HULIN

*ENSTA, LOA, Ecole Polytechnique, F-91128 Palaiseau Cedex, France
and GPS, Ecole Normale Supérieure, 24, rue Lhomond,
F-75005 Paris, France*

Resume - On passe en revue l'effet Stark optique sur les excitons dans les puits quantiques du type GaAs. Cet effet a ete observe aussi bien dans le cas d'un couplage dynamique entre etat fondamental du cristal et premier niveau excitonique que pour un couplage entre les deux premieres bandes quantifiees de conduction.

Abstract - The optical Stark shift of excitons in GaAs and related compounds is reviewed. It has been observed for a dynamic coupling between either the crystal ground state and first exciton or between the two conduction-band sublevels.

Irradiation of a medium with non-resonant light leads to a shift of the energy levels of the system. This optical, or dynamical Stark effect has been studied for may years in atomic gases, where narrow transition linewidths make its detection relatively easy. It is only recently however, that it could be detected for excitonic transitions in crystals.[1,2,3,4] This development has stirred considerable interest, because the optical Stark effect has inherent features which makes it well-suited for applications such as in ultrafast all-optical data processing. Since the shift-inducing pump beam falls in a transparency region of the medium, it experiences little loss, in contrast to optical non-linearities based on a saturation mechanism which requires dissipation of pump beam energy. Also, because virtual transitions are involved, the light-induced shift of exciton levels follows closely the pump pulse duration, making it possible to design ultrafast all-optical logic gates with subpicosecond switch-on and switch-off times.

The first observation of a resonant optical Stark effect in a semiconductor was reported by Fröhlich et al.[1] in Cu_2O. The pump beam photon energy was set close to the transition between 1S and 2P excitons of the yellow series. A weak test beam measured the ground to 2P exciton transition energy. In the presence of the pump pulse, a splitting and small shift of the exciton level was observed.

This excited state optical Stark effect has to be contrasted with the dynamical coupling between ground and first exciton which was observed shortly thereafter in GaAs quantum wells.[2,3] Using femtosecond time-resolved detection techniques, Mysyrowicz et al.[2] demonstrated very large light-induced displacements of the lowest heavy-hole exciton resonance energy, up to 8 meV, a substantial fraction of the exciton binding energy itself. Moreover, it was verified that the effect is very rapid, essentially following the subpicosecond pump pulse. Von Lehmen et al.[3] have measured the amount of shift as a function of detuning and pump intensity, also in GaAs quantum wells. More recently, K. Tai et al.[4] have observed the optical Stark effect in InGaAs/InP at room temperature. This last system is particularly interesting for applications since the lowest exciton is located at 1.5 μm, an optimum wavelength for transmission in optical fibers. Finally, in a very recent development, Fröhlich et al.[5] have measured the shift resulting from a dynamical coupling between the two lowest conduction-band sublevels in a GaAs quantum well structure.

Before going into more details on these results, we briefly summarize the different theoretical models developed so far. A first approach consists in applying to excitons the dressed atom picture. The dressed atom picture has proved very powerful in describing the optical Stark shift and related effects in atomic systems.[6] This model usually restricts itself to a two-level system, an approximation valid for near resonant conditions. In contrast to usual treatments of nonlinearities based on a perturbation expansion in successive orders of the pump field, it solves the interaction between atoms (excitons) and the radiation exactly to all orders. For excitons it predicts a shift

$$\delta E = ((E_x - \hbar\omega_p)^2 + 4|\mu E_p|^2)^{1/2} - (E_x - \hbar\omega_p) \tag{1}$$

or, for $(E_x - \hbar\omega_p) > 4|\mu E_p|$

$$\delta E \simeq \frac{2|\mu E_p|^2}{E_x - \hbar\omega_p} \qquad (2)$$

where μ is the transition matrix element between ground and exciton. E_x is the exciton energy and $\hbar\omega_p$ the pump photon energy.

Thus, the intense pump beam "repels" the energy levels, leading to a blue shift for positive detuning $E_x - \hbar\omega_p > 0$ and to a red shift for $E_x - \hbar\omega_p < 0$.

The dressed exciton model, although in good quantitative agreement with experiments (see below), does not predict another concomitant effect, well apparent in fig. 1), namely a reduction of exciton oscillator strength. This last effect has its root in the composite nature of excitons, (electron and hole) and requires a more elaborate treatment which takes into account the Fermi nature of the constituents. Such a model, with the many-body interactions between excitons treated from first principles has been derived by Schmitt-Rink et al.[7] Here, the coherent polarization of the medium driven by the external pump field is viewed as an artificial condensate of excitons. Since the particles are only virtually excited, the system evolves in the absence of thermal excitations (T = 0). The weak test beam, by adding a few particles to the system, effectively probes the collective excitation spectrum of the pseudo-condensate. Drawing from similar results obtain by Keldysh and Kozlov,[8] and Nozieres and Comte,[9] this theory covers in a unified manner both the low density (weak interaction) limit with real-space pairing of electron-holes (Bose condensation of tightly bound excitons) and the high density, strong interaction limit where momentum pairing prevails (condensation of electron-hole Cooper pairs). Although conceptually very appealing and in principle exact, this treatment requires formidable computations. For application to specific cases, such as GaAs quantum wells, Schmitt-Rink and Chemla[7] propose an approximation which reduces the collective excitation spectrum to the well-known Bogoliubov dispersion near $K = 0$ of a weakly interacting Bose gas. In the limit $na_0^3 \ll 1$ ($na_0^2 \ll 1$ in two dimensions) where n is the density of virtually excited exciton, given by:

$$n = \frac{2|\phi(r=0)|^2|\mu E_p|^2}{(E_x - \hbar\omega_p)^2}$$

one recovers equation 1), except for a correcting factor of the order unity. However, as already stated before, this approach has the merit to incorporate the exciton bleaching, which is predicted to be directly proportional to the blue shift δE.

Finally, in a slightly different context, Saikan et al.[10] have calculated in great details the inverse Raman band shape in the vicinity of a single photon resonance including the case of an optical Stark shift between excited levels. These theoretical results can be transferred directly to the case of a dynamical coupling between excited states of a semiconductor.

So far, all experimental results are consistent with theory. The light-induced shifts of the lowest heavy-hole exciton in GaAs/AlGaAs or InGaAs/InP quantum wells are linear with pump intensity I_p for $I_p \lesssim 10^8 W/cm^2$ with a slope given by equation 2, within the uncertainty (factor 4) in the determination of the laser intensity. At higher intensities $I_p \gtrsim 10^9 W/cm^2$, however, a departure from linearity is observed in GaAs quantum wells. This effect is not predicted in the dressed atom model or the low density limit Schmitt-Rink model. It finds its explanation in the complete many-body theory.[7] The departure from linearity signals the onset of the transition from the regime of tightly bound excitons to that of extended, Coulomb correlated Cooper pairs. As mentioned before, the normalized reduction of exciton oscillator, due to phase space filling is expected to scale linearly with the normalized exciton blue shift. By taking this effect into account one obtains a good fit to the datas (see fig. 2).

An interesting question, still to be answered fully, concerns the polarization dependence of the optical Stark effect. In the experiment by Fröhlich et al. in GaAs quantum wells, the polarization of the pump beam is set either parallel or perpendicular to the quantum-well-structure growth axis c. Only for the parallel configuration is the dynamical coupling between the two lowest conduction band sublevels expected to be effective. This is corroborated by the experiment: a much larger Stark shift is observed if $\vec{E}_p \| c$ as compared to $\vec{E}_p \perp c$.

Fröhlich et al.[5] pointed out that a similar polarization dependence of the Stark effect should also occur in the perpendicular configuration $(\vec{E}_p \perp c)$ by comparing cases $\vec{E}_p \| \vec{E}_s$ and $\vec{E}_p \perp \vec{E}_s$, where E_s is the probe beam polarization vector. However no measurable difference is observed (neither for a dynamical coupling

between conduction band sublevels[5] nor for a coupling between ground and first exciton).[2] This suggests the occurence of a very fast dephasing mechanism acting on the probe excitons, of the order $T_2 \sim 300fs$ or less. As discussed by Schultheiss et al.[11] several mechanisms may conspire to give such fast dephasing rates: collisions amongst the injected probe excitons (as well as between probe free carriers and excitons) scattering of probe excitons by longitudinal acoustic phonons, a process enhanced in the experimental conditions of ref. 2) by the finite lattice temperature $T = 15K$ and by the narrow well thickness.[12] Further experiments are required to fully answer these interesting questions.

In conclusion, the basic processes responsible for the optical Stark effect in quantum wells are now understood. The further study of this effect may bring important informations on the physical properties of the system under study. An example is provided by the optical Stark effect between excited states in GaAs quantum wells,[5] where accurate values of the dipole moment and energy interval between conduction band sublevels are extracted from experiments. The study of the optical Stark effect may also bring invaluable informations on the loss of coherence of excitons in semiconductors.
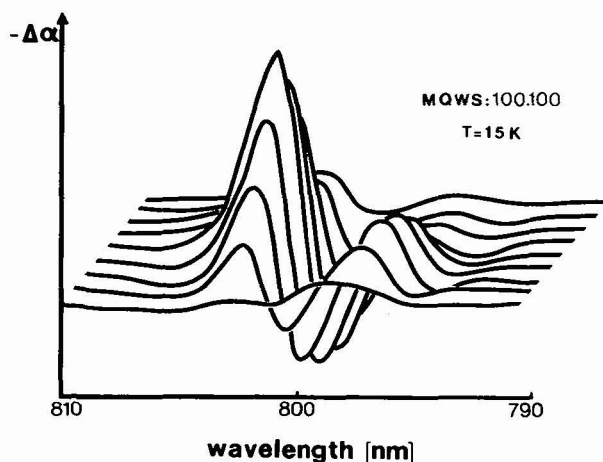

Figure 1 : Differential transmission spectra of a GaAs multiquantum-well-structure in the vicinity of the lowest (heavy-hole) exciton, recorded at 100 Fs intervals, well thickness = 100 Å ; barrier thickness = 100 Å. Pump wavelength = 815 nm.
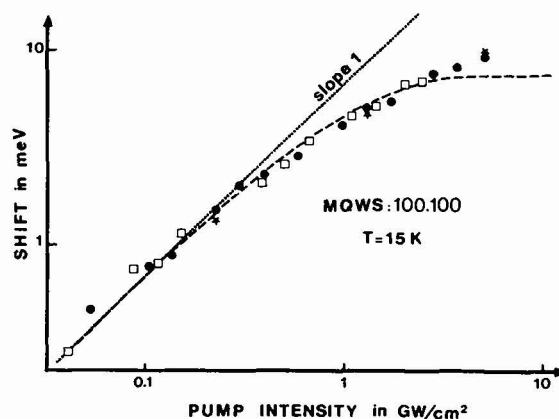
Figure 2 : Blue shift of the heavy-hole exciton as a function of pump intensity at 815 nm. The dashed line is the calculated dependence taking the bleaching of the exciton into account.

REFERENCES

1) D. Fröhlich, A. Nöhte, K. Reimann, Phys. Rev. Lett. *55* 1335 (1985).

2) A. Mysyrowicz, D. Hulin, A. Antonetti, A. Migus, W. T. Masselink, H. Morkoc, Phys. Rev. Lett. *56* 2748 (1986).

3) A. Von Lehmen, D. S. Chemla, G. E. Zucker, G. P. Heritage Optics Lett. *11*, 609 (1986).

4) K. Tai, G. Hegarty, W. T. Tsang, Appl. Phys. Lett. *51*, 152 (1987).

5) D. Fröhlich, R. Wille, W. Schlapp, G. Weimann, Phys. Rev. Lett. *59*, 1748 (1987).

6) C. Cohen-Tannoudji, S. Reynaud in Multi-Photon Processes, G. Eberly editor (Wiley, New York 1977).

7) S. Schmitt-Rink, D. S. Chemla, Phys. Rev. Lett. *57*, 2752 (1986).

8) L. V. Keldysh, A. N. Kozlov, Sov. Phys. JETP, *27*, 571 (1968).

9) P. Nozieres, C. Comte, J. de Physique *43*, 1083 ((1982).

10) S. Saikan, N. Hashimoto, T. Kushida, K. Namba, J. Chem. Phys. *82* 5409 (1985).

11) L. Schultheis, J. Kuhl, A. Honold, C. W. Tu, Phys. Rev. Lett. *57*, 1635 (1986) *57*, 1797 (1986).

12) T. Takagahara, Phys. Rev. B *32*, 7013 (1985).