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**FEMTOSECOND STUDY OF THE OPTICAL STARK EFFECT IN MULTIPLE QUANTUM WELL STRUCTURES**

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**Résumé** Les résonances excitoniques dans les structures à puits quantiques multiples peuvent être fortement modifiées par une excitation optique dans la zone spectrale de transparence. Cet effet Stark optique les déplace vers les plus hautes énergies tout en diminuant leur intensité jusqu'à leur disparition quasi-complète. Une relation linéaire existe entre ces deux effets. Le déplacement des raies n'est pas linéaire avec l'intensité lumineuse aux fortes excitations mais ce comportement s'explique en tenant compte de la réduction simultanée de la force d'oscillateur.

**Abstract** Excitonic resonances in MQWS can be strongly affected by exciting the semiconductor in the transparency region. This optical Stark effect shows up as a blue shift of the excitonic lines associated with a reduction of the oscillator strength and is apparent only while the electromagnetic field is present. We have studied the dependence of this shift as a function of intensity and showed in particular how its saturation is related to the oscillator strength reduction.

Excitonic resonances are at the origin of large optical non-linearities, especially in Multiple Quantum Wells Structures (MQWS) where moreover they are present even at room temperature. These features make MQWS very attractive both from a fundamental point of view and for applications to optical devices. The recently reported optical Stark effect [1,2] has raised a wide current of interest since it corresponds to the coupling of electronic states with photons of energy below the absorption edge. The use of a pump wavelength in the transparency region of the MQWS offers great promise for optical devices since it implies an ultrafast response time and almost the absence of heat dissipation. It was already known for atoms that the virtual absorption and re-emission of non-resonant photons leads to a shift of the atomic levels. In solids such a situation should also be encountered for transitions like excitons. The small magnitude of the corresponding energy shift with respect to typical excitonic transition linewidth requires the use of lasers with quite high peak power. This is obtained by using very short laser pulses ( $10^{-13}$  s) which in the same time yield direct informations on the dynamics of this optical Stark effect. In our experimental conditions, the magnitude of the effect can be large. We report here its study in function of the laser intensity.

The experiments are performed following the classical pump and probe scheme. The primary laser source is the amplified output of a colliding-pulse mode-locked dye laser. The pulse duration is shorter than 100 fs with an energy per pulse of 0.3 mJ at 620 nm and 10 Hz repetition rate. The beam is split in two parts, both of them being focused on two different water cells to produce two wavelength continuums of 100 fs duration. The pump pulse is obtained by a selection inside the continuum of the appropriate wavelength with an interferential filter and a further amplification in a double-pass amplifier pumped by a frequency-doubled YAG laser. The filter spectral width implies a pump duration of 400 fs leading to an incident maximal power of 5 GW/cm<sup>2</sup> on the sample. The probe beam is obtained by strongly attenuating the continuum. The polarization of the probe is set parallel or perpendicular to that of the pump. The sample is a GaAs-AlGaAs MQWS with 100 Å thick wells and barriers and is held at T = 15K. The spectrum of the probe pulse transmitted through the pump-irradiated sample is recorded at different delays between the pump and the probe on an optical multichannel analyzer.

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Absorption spectra recorded at the maximum of the pump pulse are displayed on figure 1 for different pump intensities. The pump wavelength 827 nm corresponds to a detuning of 46 meV with respect to the lowest absorption line, i.e. the heavy-hole (h-h) exciton (802.5 nm). The most striking feature in the excitonic resonances modification lies in the evolution towards higher energy (blue shift) when the intensity of the pump increases. A shift of 8 meV and more can be obtained for the h-h exciton, indicating that the effect can be quite large and is not only a small modulation. The light-hole (l-h) exciton is also affected but in a milder way; there is a factor 3 in the magnitude of the energy shift for the h-h and l-h excitons. The other noticeable feature in figure 1 is the decrease of the exciton oscillator strength simultaneous with the line shift. For the highest intensity, an almost complete disappearance of the h-h line occurs, making it

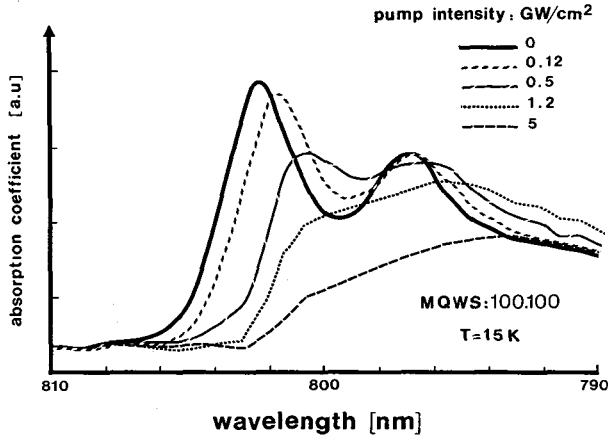


Figure 1 Time-resolved absorption spectra of a MQWS at 15 K during the presence of an intense optical pump pulse at 827 nm. The wells and the barriers are both of 100 Å width. The pump intensities are as indicated.

difficult to measure the shift in this extreme case. The observed broadening is not very large and may partly be due to some fluctuations of the pump intensity although a small window was applied to select pump pulses with energies within 20% variation. The overall effect is also smaller for the l-h exciton. After the end of the pump pulse (situation not shown on the figure), the recovery of the absorption spectrum is almost complete. The excitonic lines are again exactly in the same position as before the excitation. Only a small reduction of the oscillator strength indicates the presence in the system of a probably small population of free electrons and holes, possibly created through a two-photon absorption process. Otherwise, no real exciton population has been built during this virtual excitation.

We have reported on figure 2 the relative variation  $\Delta f$  of the oscillator strength  $f$  in function of the observed energy shift  $\Delta E$  relative to the 2-dimensional exciton binding energy  $E_x$ . The experimental points follow quite well a linear law:

$$\Delta f / f_0 = \Delta E / E_x \quad (1)$$

except for the highest intensity where the small amplitude of the excitonic line may lead to some uncertainty. This result should be compared with the relationship between  $\Delta f/f$  and  $\Delta E/E_x$  in MQWS in presence of a real population of excitons [3]. The same law has been found but for wells of 50 Å thickness. In the case of the 100 Å thick wells (same sample), no appreciable blue shift could be observed due to the strong reduction of the oscillator strength in presence of a large number of excitons.

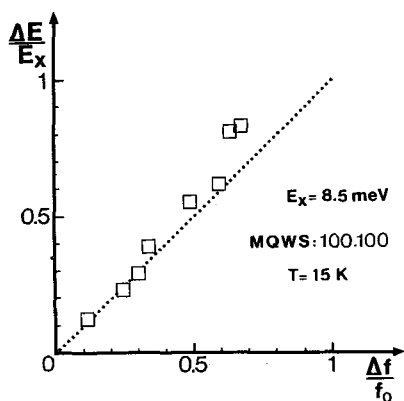


Figure 2 Relative h-h exciton energy shift in function of the relative decrease of the exciton oscillator strength.

By transposing the picture of dressed atoms to excitons, the model of the dressed exciton [1] predicts when the pump beam is in the transparency region of the crystal, a blue shift  $\Delta E$  of the exciton line given by:

$$\Delta E = \hbar \sqrt{(\omega_x - \omega_p)^2 + \Omega^2} - \hbar (\omega_x - \omega_p) \quad (2)$$

$\hbar \omega_x$  and  $\hbar \omega_p$  are the exciton and pump photon energies.  $\Omega$  is the Rabi frequency and its square is proportional to the intensity of the electromagnetic field and the oscillator strength of the excitonic transition. At low pump intensity, expression (2) can be approximated by:

$$\Delta E = \Omega^2 / 2\hbar (\omega_x - \omega_p) = a I f / (\omega_x - \omega_p) \quad (3)$$

where  $a$  is a constant. This has to be compared with a recent calculation of S. Schmitt-Rink et al.[4] where at low pump intensity the shift has a similar expression:

$$\Delta E = 2|\mu E_p|^2 |\phi_{1s}(r=0)|^2 / \hbar(\omega_x - \omega_p) N_S$$

where  $\mu$  is the interband dipole matrix element,  $E_p$  the pump field,  $\phi_{1s}$  the wave function of the  $1s$  exciton and  $N_S$  the saturation density due to excitonic phase-space filling. In both cases, the blue shift is proportional to the pump intensity, the exciton oscillator strength and the inverse of the detuning, the difference lying in the proportionality factor.

We have reported in figure 3 the magnitude of the measured blue shift in function of the pump intensity for the heavy-hole exciton. The detuning between the pump and the h-h exciton is 46 meV. At low pump intensity, the predicted linear behavior is observed. However, for intensities larger than  $0.3 \text{ GW/cm}^2$ , the shift starts to saturate and never go beyond the exciton binding energy. This departure from the linear dependence may have different origins:

- i) in the model of the dressed exciton, the pump intensity appears under a square root and the linear approximation is valid only if the Rabi frequency is smaller than the detuning. The strongest observed blue shift corresponds to  $\hbar \Omega = 26 \text{ meV}$ ; this term is not negligible with respect to the detuning but the error introduced by using the approximation (3) instead of the square root law is only 10% and cannot be responsible for the energy shift saturation;
- ii) in the expression (3) the oscillator strength of the excitonic transition appears in the numerator which depends itself of the magnitude of the electromagnetic field. For an increase of the pump intensity from  $I$  to  $I + dI$ , the corresponding

increase of the excitonic shift has not to be calculated on the unperturbed oscillator strength  $f_0$  but on the value  $f$  already perturbed by the field intensity  $I$ . By using the experimentally determined linear relationship between  $\Delta E$  and  $\Delta f$ , one obtains:

$$\Delta E = E_x [1 - \exp(-a f_0 I / E_x \Delta \omega)] \quad (4)$$

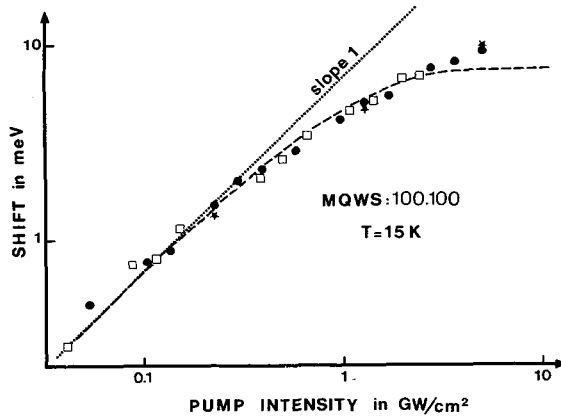


Figure 3 Variation of the h-h exciton energy in function of the pump intensity on a log-log scale. The dotted line corresponds to the linear law, the dashed line to expression (4) in the text.

The dashed line on figure 3 is the plot of this expression and as can be seen, fits quite well with the experimental points. We conclude therefore that the dependence of the oscillator strength with the pump intensity is itself sufficient here to explain the observed saturation of the energy shift.

To summarize, the optical Stark effect can affect strongly the excitonic transitions. It manifests itself both as a shift towards higher energies of the exciton line and as a reduction of the excitonic oscillator strength. We have found a linear relationship between these two effects. By noting that the optical Stark effect can be considered as a virtual creation of excitons during the pump pulse, it is not too surprising to find the same type of behavior than in the case of a real population of excitons in MQWS. The major difference lies in the fact that this effect was observed in the real case only for strictly 2-dimensional systems (less than 100 Å wells), a condition not required in case of virtual excitation. Finally we have observed a non-linear dependence of the energy shift in function of the pump intensity which, we suggest, is explained by taking into account self-consistently the reduction of the oscillator strength.

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