"Dressed Excitons" in a Multiple-Quantum-Well Structure: Evidence for an Optical Stark Effect with Femtosecond Response Time

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A very large high-energy shift of exciton resonances in GaAs multiple-quantum-well structures is observed during irradiation of the sample with femtosecond nonresonant radiation. This effect is discussed in terms of excitons "dressed" by photons.

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It is well known that the physical properties of atoms can be modified by their interaction with electromagnetic radiation.[1] For instance, the virtual emission and reabsorption of nonresonant photons leads to a shift of the atomic energy levels. This effect occurs either spontaneously (in the absence of a light source) leading to the Lamb shift or it can be induced by external light, in which case it corresponds to the "light shift" or optical Stark effect.[2]

The light shift of atoms has been observed and discussed in great detail in the radiofrequency, microwave, and optical domains.[3,4] Its physical description is particularly transparent in the formalism of the "dressed atom,"[5] which incorporates in a natural manner other effects occurring for photons nearly at resonance with a transition of the system, such as an optically induced splitting of atomic lines (Autler-Townes effect)[6] or the appearance of optical gain in the absence of population inversion.[7]

In principle, a similar situation should also be encountered in solids, for intrinsic transitions like excitons. Unfortunately, experimental verification in these systems is not easily achieved because the magnitude of the effects is usually small compared with the linewidth of typical excitonic transitions. In fact, except for a recent report by Fröhlich, Nöhte, and Reimann[8] of an optically induced mixing of $1S$ and $2P$ excitons in Cu_2O accompanied by a small shift, there has been so far no evidence of light-induced exciton shifts in the literature.

In this Letter we present experimental evidence for a light shift of the lowest excitons in a GaAs-AlGaAs multiple-quantum-well structure (MQWS) induced by a light source falling in the transparency region of the medium. The use of an intense femtosecond laser source allows one to induce very large displacements of these excitons. By time resolution of the process, the virtual nature of the optical transitions involved is clearly revealed. To interpret the results, we apply the picture of the dressed atom to the case of excitons. Despite its drastic assumptions, this model explains in a simple manner many of the principal results. The occurrence of large shifts with instantaneous response times may have practical relevance for the construction of ultrafast all-optical devices such as bistable or logic elements with on *and* off switching times in the picosecond or even subpicosecond domain.[9]

In our experiment, two tunable optical pulses of femtosecond duration are used. The intense pump pulse is obtained by selection with an interference filter of a narrow frequency range from a subpicosecond white continuum. The white continuum is generated in a cell of water by focusing of the amplified output of a 150-fs colliding-pulse mode-locked laser. Typical intensities of the pump pulse at the sample surface after further amplification in a one-stage dye amplifier are in the range 10^9–10^{10} W/cm^2. The much weaker probe pulse is given by the broad-band emission generated in a different water cell. The spectrum of the probe pulse transmitted through the pump-irradiated sample is recorded at different delays between pump and probe pulses, yielding a subpicosecond time-resolved picture of the absorption of the sample. The polarization vector of the probe has been set either parallel or perpendicular to that of the pump, with identical results in both cases. We have studied several MQWS samples held at 15 K having different well and barrier sizes. Qualitatively similar results have been observed in all samples.

Absorption spectra representative of the pump-pulse photon energy tuned well below the lowest heavy-hole exciton are shown in Fig. 1 for a MQWS having 100 Å well and barrier sizes. As can be seen, a very large blue shift of the exciton is observed *during* the presence of the pump optical pulse. The fact that the sample returns to its original state after the end of the excitation shows that no substantial population of excitons or free carriers has been generated by the pump

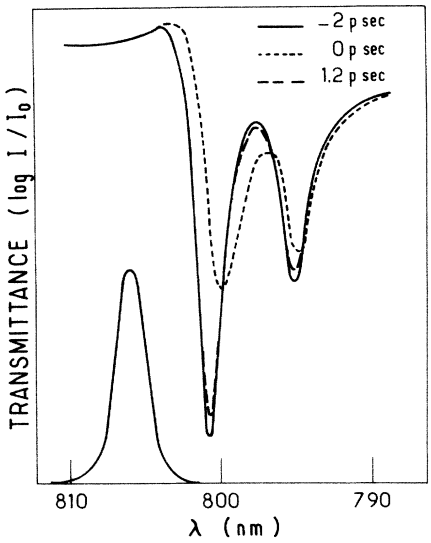

FIG. 1. Subpicosecond time-resolved absorption spectrum of a GaAs-AlGaAs MQWS sample with well and barrier thickness of 100 Å, recorded at different delays from a nonresonant intense optical pump pulse. The spectral position of the pump pulse is also shown in the figure. Sample temperature is 15 K.

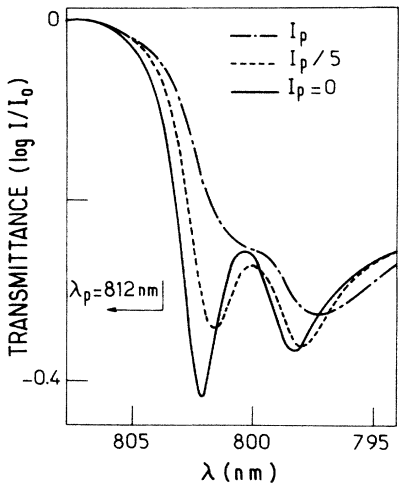

FIG. 2. Subpicosecond time-resolved absorption of a MQWS (GaAs well thickness 100 Å; AlGaAs barrier 25 Å) at 15 K, for different pump-pulse intensities. The pump-pulse wavelength is 812 nm. Absorption spectra have been recorded at the time of maximum of the pump pulse.

pulse, as expected from energy conservation.[10] In Fig. 2, the modifications of the absorption spectrum of a different MQWS (100-Å-thick well, 25-Å-thick barrier) are shown as a function of the pump intensity for a fixed time delay set at the maximum of the pump pulse. A very large blue shift is visible both for the heavy-hole and light-hole excitons (heavy hole at 802 nm, light hole at 798 nm) but is more pronounced for the heavy-hole exciton. In fact, the field-induced effect on the heavy-hole exciton is so large that it results in an almost complete disappearance of the exciton oscillator strength at the largest pump intensity. Here again, the system recovers to its unperturbed state after the end of the pump pulse. Note that in Fig. 2 the pump wavelength is far away from the resonance, lying outside the wavelength scale of the figure.

In Fig. 3, a quasiresonant situation is shown (MQWS of 40-Å-wide well, 100-Å-wide barrier), with the pump pulse now tuned inside the low-energy tail of the inhomogeneous exciton line. The time delay of 0 psec corresponds to the maximum of the pump pulse. Note the strongly distorted exciton absorption, with an absorption peak shifted to higher energies, and the appearance of *optical gain* at lower energies from the pump pulse. The spectral position of the narrow peak in the optical gain does not correspond to a reported exciton decay mechanism. At later times, the narrow gain evolves rapidly towards a broader structure located in a region of luminescence. Note also that, in contrast to the cases shown in Figs. 1 and 2, changes in the absorption spectrum subsist after the end of the pump. These long-lasting changes, associated with a real population of excitons generated by the pump pulse, have been discussed in previous publications.[11-13]

In order to explain these results, we apply the dressed-atom picture to excitons. Consider the crystal ground state in the presence of n photons. The corresponding state $|0,n\rangle$ is energetically nearly degenerate with the state $|x,n-1\rangle$ where one photon is removed

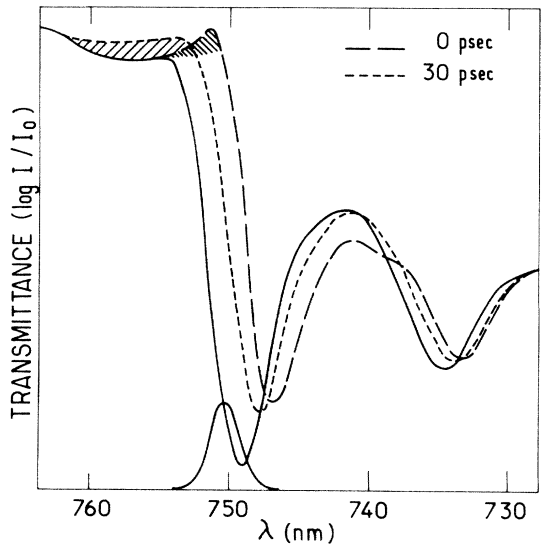

FIG. 3. Time-resolved absorption spectrum of a MQWS (GaAs well = 40 Å; AlGaAs barrier = 100 Å) at 15 K recorded during and 30 ps after a quasiresonant pump pulse at 7502 Å. Cross-hatched regions correspond to optical gain. Maximum of short-lived gain is at 7515 Å.

and one exciton is created. A similar situation holds for states $|0,n+1\rangle$ and $|x,n\rangle$, and so on. Applying the exciton–radiation-field interaction between these nearly degenerate states leads (see Fig. 4) to new mixed eigenstates α_n, β_n; α_{n+1}, β_{n+1}; etc., which are split by the quantity

$$\hbar\Delta=\hbar[(\omega-\omega_0)^2+\Omega^2]^{1/2}, \quad (1)$$

where $\omega-\omega_0$ is the detuning of the pump laser frequency ω from the exciton resonance at ω_0, and Ω is the Rabi precession frequency

$$\hbar\Omega=p_{0x}|E|, \quad (2)$$

with $E^2/8\pi=n\hbar\omega/c$ and p_{0x} the effective matrix element between ground and exciton. It can be shown that for negative detuning ($\omega-\omega_0<0;|\omega-\omega_0|>\Gamma$, where Γ is the exciton linewidth), essentially only the lower-lying levels α_i are occupied. Corresponding optical transitions, probed by a weaker beam, are shown in Fig. 4.

This picture of excitons dressed by the pump photons explains well the results of Fig. 1. The blue-shifted "exciton" absorption corresponds then to transitions $\alpha_n\rightarrow\beta_{n+1}$. The magnitude of the observed exciton shift is compatible with that derived from expression (1) although a precise comparison is not possible in view of the large uncertainty in the estimate of the pump field strength E. We have verified that in our experimental conditions, the term Ω in (1) is important even for large detuning, by measuring a weakly dependent exciton blue shift as a function of detuning at constant driving field strength.

We can apply the same model to the resonant situation, in order to interpret the observed short-lived amplification as well as the blue-shifted absorption. According to Ref. 7, a down-cascading process $\alpha_i\rightarrow\beta_{i-1}$ leads to optical gain which has a peak efficiency of a few percent for $\hbar(\omega-\omega_0)=3\Gamma$ and which becomes negligible at larger detunings. We suggest that the short-lived amplification observed in the early times in a narrow spectral region lying almost symmetrically to the upshifted exciton with respect to the pump beam frequency corresponds to this process (see Fig. 3).[14] As mentioned above, this narrow amplification rapidly evolves towards a broader gain in the region of luminescence. A more detailed study of this evolution which could bring useful information upon the loss of coherence of the excited system is in progress.

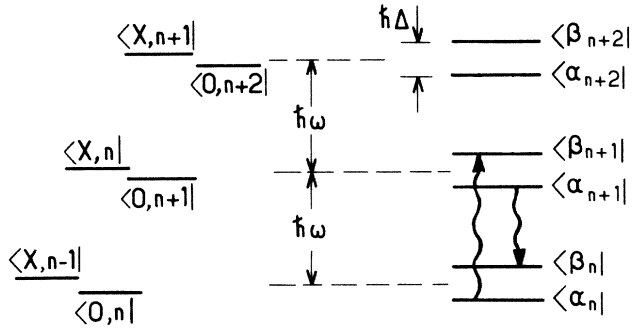

FIG. 4. Schematic representation of relevant energy levels without (left) and with exciton photon interaction (right). Upwards (downwards) arrows correspond to blue-shifted exciton absorption (red-shifted gain).

Of course, in view of its drastic assumptions, our simplified model cannot provide a detailed interpretation of all observed phenomena. For instance, it predicts complete bleaching at exact resonance, as a result of equal populations in levels α_i and β_i, which results in a complete balance between absorption and emission processes. Experimentally, only partial bleaching of the absorption is observed. But the tacit assumption of a homogeneously broadened line is unrealistic and becomes certainly inadequate in a resonant situation. Also, a red shift of the exciton should be observed, for a pump laser tuned to the high-energy side of the resonance, a behavior which is not observed experimentally. However, the restriction of the discussion to a two-level system is then incorrect. Higher-energy-lying excitons as well as the ionization continuum should be taken into account in that case.

As a last brief comment, we wish to point out that the "exciton light shift" reported here does not appear to be sensitive to the size of the well, in contrast to the long-lasting exciton blue shift associated with a real population of excitons at high densities.[12] Indeed, we have also observed an optical Stark effect of excitons in a 0.7-μm-thick GaAs sample. This suggests that a similar effect may be expected in a wide variety of semiconductors.

To summarize, we have reported the first evidence of very large displacements of excitonic resonances in MQWS induced by nonresonant radiation. This effect has been interpreted within the framework of the "dressed exciton" model. The same model can be applied to a simple explanation of a short-lived optical gain observed under near-resonant pumping. We anticipate future applications of this effect in ultrafast all-optical switching.

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[1]For a recent review, see, for instance, C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, "Photons and Atoms: Introduction to Quantum Electrodynamics" (Wiley,

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[2]The term "lamp shift" was coined by A. Kastler to draw the parallel with the Lamb shift.

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[9]For a recent review see, for instance, *Optical Bistability III*, edited by H. M. Gibbs *et al.*, Springer Proceedings in Physics Vol. 8 (Springer-Verlag, New York, 1986).

[10]The very small difference in the spectra recorded before and 1.2 ps after the pump pulse (see Fig. 1) is probably induced by some carriers created either by a two-photon absorption process or by linear absorption of a small residual high-energy tail of the pump pulse.

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[14]It may be worth mentioning that together with transient optical gain a splitting of the exciton fluorescence line is predicted during the presence of the pump field (see Refs. 3 and 6).