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PROPAGATION EFFECTS AND OPTICAL NONLINEARITIES OF EXCITONS IN MULTIPLE QUANTUM-WELLS

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RESUME : Nous avons étudié les mécanismes qui sont à l'origine des nonlinéarités optiques excitoniques dans les Multipuits Quantiques GaAs/GaAlAs. L'analyse des spectres obtenus révèle, outre les nonlinéarités attendues, des effets propagatifs fortement dépendent de la température. Ces résultats mettent en avant le rôle de la propagation de la lumière à travers un milieu non-linéaire.

ABSTRACT : We have investigated the mechanisms that give rise to the optical nonlinear response associated with excitons in 2D GaAs/GaAlAs Multiple Quantum-Well (MQW) structures. The analysis of the spectra has revealed in addition to the expected sources of excitonic optical nonlinearity, temperature dependent propagation effects. These experiments bring to the fore the role of light propagation through a nonlinear medium.

INTRODUCTION

The large room-temperature optical nonlinearities of excitons in GaAs/GaAlAs Multiple-Quantum Wells (MQW) have elicited a lot of interest in recent years. However, the detailed interpretation of nonlinear optical experiments on MQW is still subject to controversy, as the macroscopic manifestations of these nonlinearities (refractive or absorptive changes) as well as their incidence on light propagation through the sample, are still not well understood.

Room-temperature nonlinear optical experiments in a 65-period thick MQW have indicated that the contribution of the heavy exciton in degenerate four wave mixing (DFWM) is much larger than that of the light exciton, while in the nonlinear transmission (NLT) spectra the peaks of the two excitons were of comparable intensities^[1]. This was attributed to the different contribution of the real and imaginary parts of the effective third-order susceptibility in the two nonlinear techniques. This conclusion, however, is at variance with experiments at liquid Helium temperatures^[2] where both DFWM and NLT give a large contribution for the heavy exciton. In addition, the difference between the room-temperature and low-temperature results has not received an adequate explanation.

In this Communication we show that propagation effects can account for the main features of all these nonlinear spectra.

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EXPERIMENTAL

Our samples are MBE-grown GaAs/Ga_{0.7}Al_{0.3}As MQW, with 120 Å-wide wells and 80 Å barriers. The MQW region is sandwiched between two 2-micron thick layers of Ga_{0.7}Al_{0.3}As. Two types of samples were used, having thicknesses of 30 and 60 periods respectively. A 2mm² hole was etched chemically in the GaAs substrate to allow optical transmission experiments in the MQW.

The light source is a parametric generator² producing 10-psec long infrared pulses of peak intensity 100 KW/cm², having a spectral width of 1 meV (0.5 nm at 800 nm). It consists of several LiIO₃ crystals (total effective length 10 cm) pumped at 532 nm by the second harmonic of a pulsed, mode-locked YAG laser. Wavelength can be scanned by angle-tuning the LiIO₃ crystals.

The output of the parametric generator is split into two beams which are recombined inside the sample. One beam (pump) is very intense so that it produces optical saturation, while the second (probe) is attenuated so as not to produce any nonlinear effects. The probe transmission through the sample is measured both in the presence and in the absence of the pump, and the two measurements are ratioed. The nonlinear spectrum thus gives the pump-induced transmission of the probe, as a function of the wavelength of the incident radiation.

Figure 1
Nonlinear Transmission Spectra
at 200 K.
(a) 60- period sample
(b) 30- period sample
Heavy exciton is at 831 nm
Light exciton is at 825 nm

Figure 2
Nonlinear Transmission Spectrum
at 10 K.
60- period sample
Heavy exciton is at 804.5 nm
Light exciton is at 800.5 nm

Figure 1 reproduces the NLT spectra of the 60- and 30- period samples at 200 K. The room temperature spectra display similar characteristics but with lower S/N ratio. In the 60-period sample the heavy and light hole exciton signals have comparable intensities while in the 30 period sample the heavy exciton is dominant. At 10 K, however, (Figure 2) the heavy exciton dominates for both the 60-

and 30-period samples. DFWM spectra of the 60-period sample were also obtained, but are not displayed here; they resemble those of Figure 2, in that they show a very strong heavy exciton.

The dominant heavy exciton contribution obtained with our samples by the DFWM and the low-temperature NLT methods is similar to the published NLT and DFWM spectra^[1,2]. In the 60-period sample at room temperature, the results are once again similar to published spectra in that the heavy and light excitons have comparable strengths (with minor differences in the lineshapes probably due to different pumping conditions: our pump intensities are 100 times that in Ref. 1). On the other hand, the 30-period room temperature spectrum, displays a completely different behaviour: the heavy exciton is very strong, comparable thus to the DFWM or the low-temperature NLT results.

This last feature points to propagation effects in the non-linear spectra, since the only physical difference between the two types of samples is that light travels twice the distance in the thick samples. These effects are discussed in the next section.

DISCUSSION

The room-temperature absorption coefficient at the peak of the heavy exciton in these MQW's^[1] is of the order of $1.2 \times 10^4 \text{ cm}^{-1}$. This implies that linear propagation of a beam entails attenuation by a factor of 4 or 5 for a 60- (or 65-) period MQW. If the overall bleaching is weak, saturation is proportional to the local intensity, and this latter quantity differs by a factor of 5 between the two ends of the sample. For the light exciton, the absorption coefficient is slightly lower, so that bleaching is probably a little more uniform. In a pump-and-probe experiment, the probe beam averages the information of all the layers it traverses. The NLT spectrum is thus averaged over the whole thickness of the sample, giving practically the same average NLT for the heavy and light excitons in an optically thick sample. The thin-sample result, on the other hand, gives the NLT spectrum with higher fidelity since these propagation effects are less important.

In DFWM the diffraction efficiency of the transient grating is proportional to the square of the local incident intensity. The DFWM signal thus originates essentially at the upstream end of the sample, since the first few layers contribute 25 times more than the last few layers. Thus, as observed experimentally, the DFWM result always resembles the thin-sample spectrum.

At low temperatures, an additional effect comes into play. The homogeneous spectral width of the excitons, becomes of the order of .1 meV^[3], much smaller than their inhomogeneous width (of the order of 3 meV). This latter quantity represents the statistical distribution of the (local) exciton resonance frequency, which in turn is due to local variations of the widths of the quantum wells^[4]. These statistical variations are distributed spatially in the sample in all three dimensions, so that when propagating along the MQW axis in a small area of the sample, many different excitonic resonance frequencies are encountered. Clearly, when the ratio of the homogeneous to the inhomogeneous widths of the excitons decreases, the probability of encountering excitons with overlapping spectra, also decreases. Thus, in the course of propagation of a light beam, in a small area of the sample, very few layers can participate in the absorption (saturation process) of a particular spectral component of the beam, when the homogeneous width of the

excitons is much smaller than their statistical distribution. The low-temperature NLT spectrum thus corresponds to a thin-sample spectrum, with a strong heavy exciton contribution.

Clearly, these propagation effects can explain the overall appearance of the nonlinear spectra. Other physical processes, however, may also come into play. For example, the relative magnitudes of the heavy and light exciton nonlinearities are certainly influenced by refractive effects in DFWM^[1], or by thermal occupation effects at low temperatures. However, in thick samples, the exact relationship of the two peaks, is masked by propagative effects. A more quantitative treatment of propagative effects will be given elsewhere.

CONCLUSION

Our experiments have brought forth the effects of propagation in the nonlinear spectroscopy of MQW, which result from the intertwining of the saturation of the excitonic resonances with the depletion of the pump upon propagation. Consideration of such propagative effects is necessary for the interpretation of nonlinear optical spectra, as well as for modelling MQW-based nonlinear optical devices.

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