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**STEADY-STATE AND TIME-RESOLVED EXCITONIC OPTICAL NONLINEARITIES IN
MBE-GROWN ZnSe**

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Abstract - Excitonic optical nonlinearities are time resolved in MBE-grown ZnSe thin films. The nonlinear index is comparable to GaAs and the recovery time is less than 100 ps.

I. Introduction

ZnSe is an interesting material for optical switching and computing applications. Thermal nonlinearities in polycrystalline ZnS and ZnSe have been employed extensively in the past for these applications.^{1,2} However, these switches have been slow and inefficient due to the thermal nature of the nonlinearity. We report large optical nonlinearities of electronic origin and their fast response time. The magnitude and dynamics of this excitonic effect are measured at various temperatures. These ZnSe films were grown by molecular beam epitaxy (MBE) on (100) GaAs substrates. The samples studied were 0.23, 0.55, and 0.68 μm thick. The substrates were removed by chemical etching.

II. Steady-State Excitonic Optical Nonlinearities in ZnSe

In the first of two experiments, we measured the magnitude of this nonlinearity using a standard pump-probe technique. The broad-band probe and single-wavelength pump beam were of ≈ 3 ns full width at half-maximum (FWHM). Both pump and probe were synchronously generated by the same nitrogen laser. The pump beam was fixed at 435 nm directly generating an electron-hole plasma. We measured the absorption coefficient as a function of probe wavelength for various pump intensities.

Figure 1a shows the experimental absorption spectra for the 0.55 μm sample at 150 K. The spectrum labeled 0, which is taken in the absence of the pump beam, exhibits a pronounced exciton peak near 450 nm.

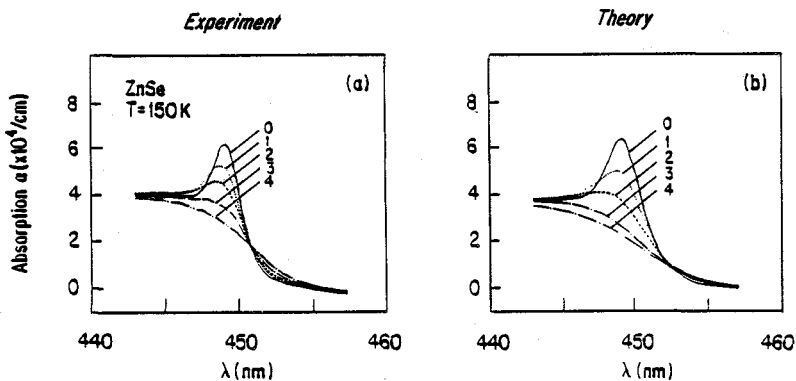


Fig. 1. (a) Experimental absorption spectra for a 0.55 μm ZnSe thin film at $T = 150$ K : 0) linear (no pump), 1) 2.5 kW/cm², 2) 9 kW/cm², 3) 23 kW/cm², 4) 46.5 kW/cm². (b) Calculated absorption spectra for different electron-hole pair densities : 0) 1×10^{15} cm⁻³, 1) 5×10^{16} cm⁻³, 2) 1×10^{17} cm⁻³, 3) 2×10^{17} cm⁻³, 4) 3×10^{17} cm⁻³

In the presence of the pump pulse, one observes a reduction of the exciton oscillator strength and a small broadening on either side of exciton. At higher pump intensities the exciton gradually disappears. This is due to the screening of the Coulomb potential by the pump-injected carriers. Similar measurements were repeated at room temperature as seen in Fig. 1b. Note, the magnitude of the nonlinear changes are comparable to those of bulk GaAs.⁵

The results were analyzed using a recently published plasma theory. This is a partly phenomenological scheme based on an extension of linear response theory with material parameters that depend on the density of the e-h plasma. This gives an expression for the nonlinear absorption spectra which takes into account screening of the Coulomb interaction, band filling, bandgap reduction, and density dependent broadening of resonances. The theoretical absorption spectra are in good agreement with the results shown in Fig.1b. The theory indicates that at room temperature the main contributions to the nonlinearities are exciton screening and density dependent broadening. At 150 K, screening of the exciton is the most significant mechanism.⁵ It has to be emphasized that in contrast to previous results on ZnSe, thermal effects do not play a role in the observed spectra.

III. Time-Resolved Excitonic Optical Nonlinearities in ZnSe

In the second set of experiments we measured the dynamics of the excitonic nonlinearities using time-resolved pump-probe spectroscopy. Free carriers were created by pumping above the ZnSe bandedge using a 310-nm pump. The ≈ 100 -fs pump pulse was obtained by frequency doubling the output from an amplified CPM dye laser. The change in transmission (ΔT) at a single wavelength was monitored by phase-sensitive detection of a continuum probe. The wavelength was tuned to the peak of the exciton and ΔT was recorded as a function of delay between pump and probe.

The exciton recovery time was investigated as a function of sample thickness, temperature, and pump intensity. The dependence on sample thickness at room temperature is shown in Fig. 2. The recovery time (τ) was faster for the thinner samples. It ranged from 110 ps for the 0.68 μm sample to 40 ps for the 0.23 μm sample. This probably results from enhanced surface recombination. τ was also seen to decrease as the temperature decreased. It decreased from 90 ps at room temperature to 50 ps at 150 K, for the 0.55- μm thick sample. We tentatively attribute this to the increased exciton oscillator strength at lower temperatures which leads to a shorter lifetime. Further investigations are in progress.

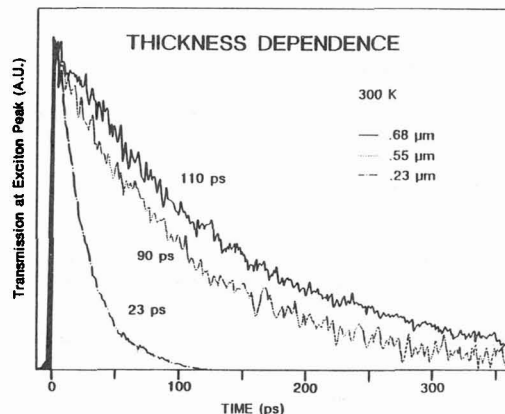


Fig. 2. Transmission at the exciton peak as a function of pump-probe delay for various thickness ZnSe thin films at room temperature.

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