Free and Bound Exciton Dynamics in Bulk II-VI Semiconductors

M. D. Martín[1], A. Amo[1], L. Viña[1], G. Karczewski[2], J. Kossut[2]

[1] *Dept. Física de Materiales, Universidad Autónoma de Madrid, Madrid, Spain*
[2] *Institute of Physics, Polish Academy of Sciences, Warsaw, Poland*

Abstract. We have studied the recombination and spin dynamics of free and bound excitons in bulk CdTe. We have found that the energy relaxation dynamics is governed by exciton-LO phonon scattering, leading to a short lived emission. Additionally, the trapping of free excitons with center of mass momentum close to zero by impurities plays a major role in the relaxation process. The circular polarization degree of the emission decreases with increasing power. However, the spin flip time remains approximately constant for all the powers used, revealing that the free exciton spin dynamics is governed by exciton localization rather than carrier-carrier scattering.

INTRODUCTION

In the last years there has been an increasing interest on CdTe, not only because this material is a suitable candidate to produce gamma and X rays detectors, but also because of the development of a new solar cell technology based on CdTe [1]. It is therefore crucial to have a precise description of the fundamental properties of this material to determine which kind of elementary excitations are responsible for the absorption/transmission/conduction of those potential devices. In this paper we present a detailed study of the recombination dynamics of excitons in high quality CdTe epilayers, together with a description of the exciton spin dynamics.

EXPERIMENT

The two samples we have studied consist of 4.5 μm thick CdTe epilayers grown by Molecular Beam Epitaxy over a ZnTe buffer layer and a GaAs substrate. The optical characterization (by means of cw photoluminescence –PL– and photoluminescence excitation –PLE–, not shown) revealed a large contribution of bound states to the PL but yet a predominant peak due to free exciton (FX) recombination. The PLE spectra show a strong coupling of excitons (X) with LO phonons, as previously reported in the literature [2]. The low temperature (5 K) emission from these two samples has been time resolved by means of a spectrograph and a syncroscan streak camera, with an overall time resolution of 10 ps. To analyze the PL into its σ^+ and σ^- polarized components we have used a combination of $\lambda/4$ plates and linear polarizers.

RESULTS

Figure 1 shows the time integrated spectrum for the smallest excitation power used in our experiments (100 μW). A clear and dominant FX peak is observed, together with several bound exciton (BX) transitions. Increasing the excitation power leads to a red shift of the emission due to many body effects [3], and under these conditions the PL becomes completely dominated by the FX recombination.

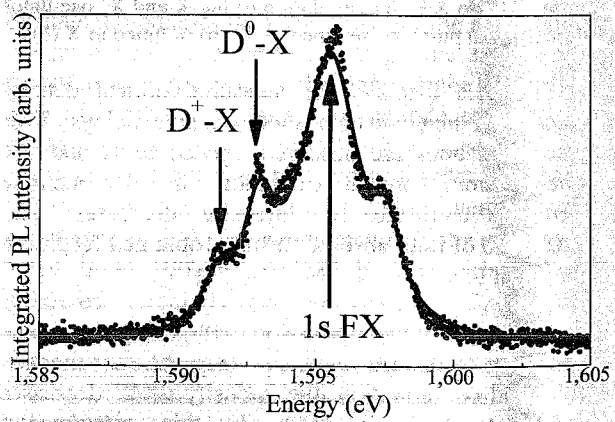

FIGURE 1. Time integrated PL spectrum for an excitation power of 100 μW.

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The time evolution of the PL for both FX and BX recombination is displayed in Figure 2. It is clear that the time to reach the maximum (t_{max}) is slightly longer for the BX (dashed line) than for the FX (solid line). Furthermore, t_{max} for the FX (inset of Fig. 2, top) shows a non-monotonic dependence on excitation power. A similar behavior has been reported recently for bulk GaAs and attributed to the competition between the capture of FX with center of mass momentum close to zero by BX states and X-X scattering [4]. Increasing the excitation power turns into a larger FX population that progressively fills the BX states. When all these states are filled, t_{max} reaches its maximum value. A further increase of the power will revert in an enhancement of X-X scattering that will accelerate the dynamics again. However, in the present case the density of localization centers is much larger than in Ref. 4 and t_{max} saturates for very high powers, hindering the observation of a re-acceleration of the dynamics through X-X scattering.

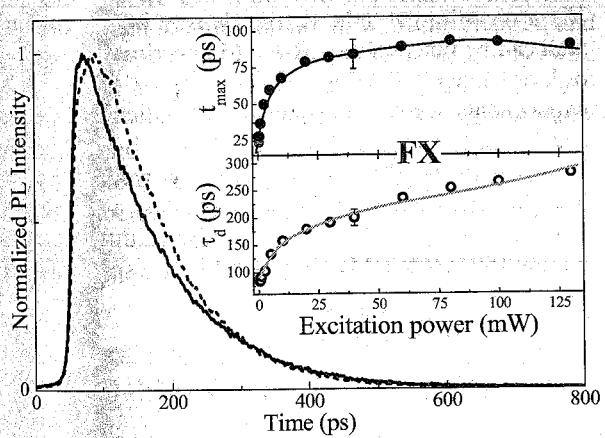

FIGURE 2. Time evolution of the PL from FX (solid line) and BX (dashed line). The inset shows t_{max} (above) and τ_d (below) for the FX emission as a function of excitation power. Lines are guides to the eye. The excitation power is 2.5 mW.

The FX PL's decay time (τ_d) (inset of Fig. 2, bottom) displays a monotonic increase with excitation power. This is again a consequence of the FX localization [5].

Figure 3 shows the maxima of the polarization degree under σ^+ excitation [$(I^+-I^-)/(I^++I^-)$, where $I^{+/-}$ denotes the $\sigma^{+/-}$ polarized emission] of the FX PL as a function of excitation power. A clear decrease with increasing power is observed. Simultaneously, the spin flip time (i.e. the decay time of the polarization degree) remains approximately constant with power (inset of Fig. 3). This fact evidences that increasing the carrier population has a negligible effect on the spin dynamics. The density of localization sites is large enough to trap most of the photocreated excitons [6] before any scattering event, through which spin relaxation will occur, can take place. Therefore, increasing the excitation power will gradually fill in more localization traps, leading to a reduction of the polarization degree of the PL, as we have experimentally observed.

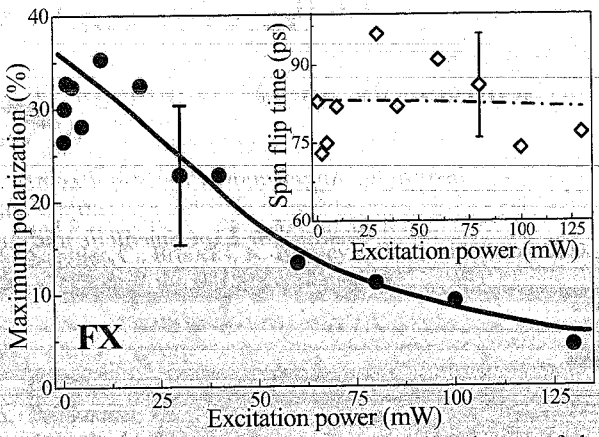

FIGURE 3. Maximum circular polarization degree of the FX PL versus excitation power. The inset displays the spin flip time extracted from a mono-exponential fit of the time evolution of the polarization. Lines are guides to the eye.

In conclusion, we have observed that the recombination and spin dynamics of excitons in high quality CdTe are governed by the localization of FX by BX states, even though the PL spectrum is dominated by FX recombination.

ACKNOWLEDGMENTS

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