Dynamics of Carriers and the Influence of the Quantum Confined Stark Effect in ZnO/ZnMgO Quantum Wells

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Abstract: We reveal the dynamics of carrier-induced screening of the internal electric field in ZnO quantum wells. By controlling the potential profile of the quantum wells we demonstrate the ability to tune the excited state lifetimes.

1 Introduction: ZnO has attracted much interest in recent years as a wide band-gap semiconductor with potential application in devices that are optically active in the blue-ultraviolet spectral region. ZnO has several advantages over its main and more established competitor, GaN. Specifically, it has an exciton binding energy of 60 meV (compared to GaN of 25 meV), a low growth temperature and low material cost [1,2].

Characterization of quantum well structures forms the basis for the development of quantum well (QW) based devices. These devices typically take advantage of quantum confinement effects, such as the ability to tune the transition energy and enhancement of the exciton binding energy. In wurtzite, c-axis grown ZnO/ZnMgO quantum wells, where the c-axis is the preferential growth direction, the mismatch of spontaneous polarizations at the well barrier interfaces results in an internal electric field across the quantum well. This electric field leads to the quantum confined Stark effect (QCSE), causes a red-shift of the transition energy and spatial separation of electrons and holes, which reduces the oscillator strength, and increases the radiative lifetime of the E1-H1 transition [2,3].

Electrons and holes generated in this polarized sate can then shield the internal field, thereby reducing the magnitude of the effective electric field and resulting QCSEs [4]. We study the evolution of the effective internal electric field through the QCSEs following excitation of carriers by an intense laser pulse of 100 fs duration. We use time-resolved photoluminescence (TRPL) to follow the changing lifetime as carriers relax and the electric field strength is restored. We also use pump-probe (PP) spectroscopy to follow the early time dynamics and the shifting transition energy. Further TRPL experiments were conducted on ZnO QWs grown with graded barriers to demonstrate an ability to exert control over the radiative lifetime of carriers in QWs that are strongly influenced by an intrinsic internal electric field.

2 Results and Discussion: TRPL (Fig. 1(a)) reveals a constantly increasing lifetime, from an initial value of 180 ns to 5.8 μs at the limits of our detection range. This was modeled by solving the Schrodinger and Poison equations self-consistently, fitting the data well. The model allows the transition energy, electron/hole overlap and lifetime to be calculated any time after excitation. Extrapolating the data beyond the resolution of our TRPL apparatus reveals a lifetime of 2 ns when the internal electric field is fully shielded and the electron-hole overlap is maximized [4].

PP spectroscopy reveals additional dynamics that occur on a much faster timescale than the 2 ns predicted. An initial decay constant of 160 ps, dependent on excitation density, was measured. These initial dynamics are attributed to either

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excitation induced effects, or the excitation density surpassing the Mott transition density. Further PP experiments where the probe is blue detuned (60 meV) from the absorption maximum of the unshielded QW reveal the dynamics of the shielding establishment. As the internal field is screened by the excited carriers, the transition is blue-shifted. In this case, the excitonic transition becomes resonant with the probe (Fig 1(b)) and we observe a transient absorption following the pump. The transient absorption is established in less than 500 fs, and is beyond the resolution of this experiment. This supports previous theory work which showed that the excitons are generated in instantaneously polarized states [5].

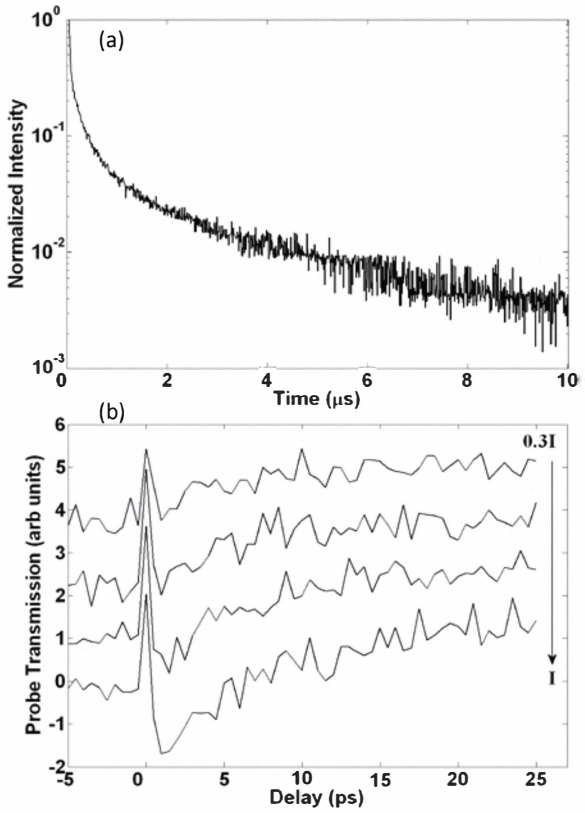

Fig. 1. (a) TRPL from 6 nm wide ZnO QW shows a decay lifetime that increases from 180 ns to 5.8 μs. (b) PP experiment probing 60 meV above equilibrium transition reveals the shielding establishment time (<500 fs).

In order to exert control over the electron-hole overlap, and hence lifetime and oscillator strength, several samples with graded barriers were proposed. These samples were realized and compared with 2 nm and 4 nm wide square QWs. PL and TRPL experiments using a streak camera were conducted. These results showed that the lifetimes and transition energies for all samples followed the trends predicted from the calculations [6]. Controlling the electron-hole wave function overlap, and hence oscillator strength, will be of great interest for optoelectronic device fabrication.

3 Conclusion: We have revealed the dynamics of carriers in ZnO quantum wells in which a large internal electric field exists. As the number of carriers increase the effective strength of the internal field is reduced because the electrons and holes are spatially separated and screen the electric field. This reduces the magnitude of the QCSE: blue shifting the transition, increasing the electron-hole overlap and reducing the radiative lifetime. The establishment of this screening is observed to occur on timescales beyond the resolution of the experiment, whilst the relaxation of the screening is clearly observed and well modeled, and allows us to predict the transition energy and lifetime for any excitation density, and at any time after excitation.

We have also revealed a means to control the intrinsic QCSE in ZnO QWs by growing samples with graded barriers. The samples grown to date have revealed increases in radiative lifetimes in agreement with model predictions. This opens the door to the growth of other samples which reduce the radiative lifetime and increases the oscillator strength over square wells for a given transition energy.

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