Investigation of carrier dynamics in $Zn_{1-x}Mg_xO$ by time-resolved photoluminescence

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ABSTRACT

The influence of the Mg concentration and lattice temperature on the carrier recombination dynamics in $Zn_{1-x}Mg_xO$ alloys has been studied by time-resolved photoluminescence for different emission and excitation energies. Carrier localization effects are found to play a significant role, becoming increasingly important for lower temperatures and higher Mg concentrations. Emission energy dependent dynamics were analyzed by the application of the theoretical model, yielding a characteristic localization energy of 60 ± 15 meV for the sample with the highest Mg concentration of $x=0.21$.

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1. Introduction

The wide-gap ZnO-based semiconductor material system shows considerable promise for the realization of optoelectronic devices in the ultraviolet (UV) spectral regime. The advantageous properties of ZnO include the direct bandgap of 3.4 eV, the large exciton binding energy of 60 meV [1,2], as well as high material quality due to improved growth methods [3–8]. Bandgap engineering for the realization of quantum confinement is achieved by alloying ZnO in ternary compounds, such as (ZnCd)O, (ZnBe)O, and, most prominently, (ZnMg)O. Recently, ZnO/(ZnMg)O heterostructures have attracted much interest as candidates for UV laser sources [9–12]. Further applications include light-emitting diodes and photodetectors. All these devices have in common that, in general, their performance is highly sensitive to the respective carrier lifetimes in the alloys.

2. Experimental details

In this work, we investigate the carrier relaxation dynamics in ternary $Zn_{1-x}Mg_xO$ alloys by time-resolved photoluminescence (TRPL). The $Zn_{1-x}Mg_xO$ samples were grown by plasma-assisted molecular beam epitaxy [13] with an average layer thickness of 0.3 μm and Mg concentrations of $x=0$, 0.04, 0.06 and 0.21. The Mg concentrations were estimated by comparing the measured PL emission energies with literature values [5,13–15] and confirmed by the X-ray diffraction. The samples were excited with a pulsed 100 fs Ti:sapphire laser at a repetition rate of 80 MHz. The pulses were frequency-tripled to a photon energy of 4.3 eV and a reference pump flux of 10^{11} photons/cm^2 per pulse. The samples were mounted inside a He-flow microscopy cryostat and the luminescence signal was collected normal to the sample surface in reflection geometry. The emission was spectrally dispersed by a 25 cm imaging spectrometer with a resolution of 0.8 nm. Subsequently, the signal was temporally resolved using a streak camera with a UV-sensitive cathode. The temporal resolution varied between 4 and 30 ps, corresponding to detection windows of 200 and 2 ns, respectively. The TRPL was measured for different lattice temperatures and at different emission wavelengths.

3. Results and discussion

As an overview, we show in Fig. 1 the PL spectra for three different $Zn_{1-x}Mg_xO$ samples in the temperature range between 10 and 290 K. The experimental data are time-integrated over the complete detection window, normalized, and vertically shifted for clarity. The sample with $x=0$ shows the well-known spectral signatures of ZnO, i.e., bound (BX) and free (FX) excitons, as well as the corresponding LO-phonon replicas [1,16]. Aside from

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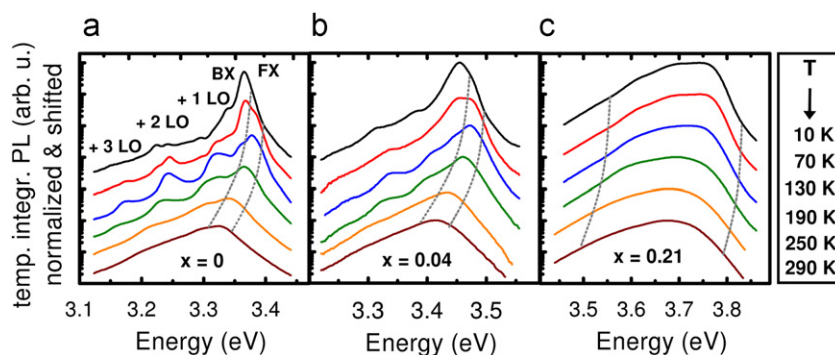E-mail address: alexej.chernikov@physik.uni-marburg.de (A. Chernikov).

Fig. 1. TRPL spectra of $Zn_{1-x}Mg_xO$ samples with Mg concentrations of $x=0$ (a) $x=0.04$ (b), and $x=0.21$ (c). The lattice temperature is varied between 10 and 290 K. The excitation energy and incident photon density are held constant at 4.3 eV and 10^{11} photons/cm², respectively. Dotted lines mark the intervals for the spectral integration.
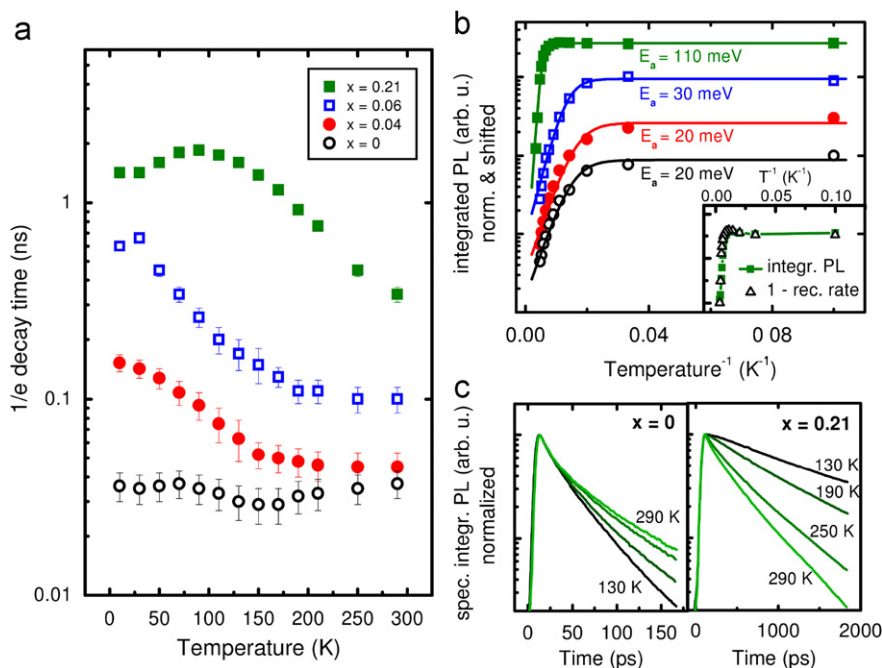

Fig. 2. (a) Temperature dependent 1/e decay times of the (ZnMg)O samples with varying Mg concentrations x. (b) Integrated PL intensities along with the corresponding fit curves. The inset shows a comparison between the integrated PL intensity for the $x=0.021$ sample and (1-recombination rate). (c) Transients of the samples with $x=0$ and 0.21 are plotted in the temperature range between 130 and 290 K.

increased broadening and a frequency shift, the spectra of the (ZnMg)O sample with a Mg concentration of $x=0.04$ exhibit spectral characteristics similar to those of the ZnO reference sample. As shown in Fig. 1(c), the broadening and the spectral shift are even more pronounced for higher Mg concentration [14].

Concentration dependent broadening of PL spectra is a common feature in ternary alloys.

It is generally attributed to spatial fluctuations of the alloy composition, such as the Mg content in the $Zn_{1-x}Mg_xO$ material system [1]. The influence of such fluctuations on the absorption and PL spectra of $Zn_{1-x}Mg_xO$ has been reported in a number of experiments [4,13,15,17]. The alloy disorder results in the formation of localized states below the band edge, known as the Urbach tail [1,18]. The presence of these states affects both the emission spectrum and the dynamics of the excited carrier system [19,20].

Fig. 2 shows the results of the TRPL measurements. To suppress the contributions of bound excitons, the transients of the $Zn_{1-x}Mg_xO$ samples with $x=0$, 0.04 and 0.06 are spectrally integrated over the PL peak with the highest emission energy, as it

is shown in Fig. 1 by the dotted lines. The data of the sample with a Mg concentration of $x=0.21$ have been spectrally integrated over the complete emission peak due to the strong spectral broadening, implying that contributions from both bound and free excitons are included.

The TRPL intensities of the ZnO reference and the 21% Mg sample are plotted in Fig. 2(c) in the temperatures range from 130 to 290 K to minimize contributions from bound excitons. The ZnO signal shows a fast decay on a 30 ps scale and a weak temperature dependence. Here, the lifetime of the excited carriers is governed by the radiative electron–hole-pair recombination and carrier capture by deep impurities [1]. In our experiments, the Auger recombination should not be of importance due to the relatively low excitation density. In contrast to the ZnO reference, the PL decay time decreases by almost one order of magnitude for the sample with $x=0.21$ as the temperature is raised from 130 to 290 K. At the same time, the absolute carrier lifetimes are significantly longer than in the pure ZnO sample, excluding a trivial explanation by faster non-radiative recombination rates in the $Zn_{0.79}Mg_{0.21}O$ sample. The temperature dependent 1/e decay