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Time-resolved photoluminescence properties of hybrids based on inorganic AlGaIn/GaN quantum wells and colloidal ZnO nanocrystals

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Abstract

Dynamic properties are studied for the AlGaIn/GaN quantum well (QW) structures with and without the coating of colloidal ZnO nanocrystals (NCs). The QW exciton recombination rate was reduced in such hybrids compared to the bare QW structure only in the sample with the thinnest cap layer of 3 nm. Assuming that one of the recombination mechanisms in this hybrid is non-radiative resonant energy transfer (NRET) between the QW and the energy acceptor material i.e. ZnO NCs, the maximum pumping efficiency was estimated to be 42% at 60 K. The NRET effect is, however, vanished after several months despite that the hybrid structures are composed of chemically stable components.

Introduction

Novel hybrid light emitting diodes (LEDs) designed to utilize non-radiative (Förster) resonant energy transfer (NRET) from excitation generated in inorganic III-N quantum wells (QW) to excitons in organic films or colloidal nanostructures might have a better efficiency compared to common hybrid LEDs [1,2]. There are several conditions, which should be satisfied in such hybrids: (i) the QW emission and the fluorescent film absorption should overlap and (ii) the interaction distance (barrier thickness) between two materials should be comparable with the QW exciton Bohr radius. The efficiency of NRET can be estimated from the quenching of the QW exciton lifetime in the presence of colloidal nanocrystals (NCs) or polyfluorene [2,3]. The bottleneck is that the degradation of polyfluorene films and/or an interface region limits the operation time of hybrid LED structures. From this point of view, it is beneficiary to fabricate such hybrids using stable NCs. The dynamic properties of the QW exciton in such hybrids have been reported previously [3]. In this work, we have studied the NRET efficiency and discuss the stability of the hybrid samples based on ZnO NCs and AlGaIn/GaN QWs designed to utilize NRET.

Experimental details

Hybrids have been based on colloidal ZnO NCs (from Sigma–Aldrich) deposited by spin coating on the top of AlGaIn/GaN QWs grown at 1050 C by metal–organic vapor phase epitaxy. The schematic drawing of the hybrid structure is shown in Fig. 1 as the inset. ZnO with the band gap of 3.3 eV plays a role of energy acceptor material while GaN with $E_g = 3.4$ eV serves as an energy donor. Optical properties of ZnO nanostructures are studied previously and will not be discussed in this work [4,5]. The AlGaIn barrier thickness was adjusted to 3 nm allowing dipole–dipole interaction in the sample. The Al content in the alloy was 16%. Similar structures, however with thicker barrier layers of 6 and 9

nm, have been grown for control. For time-resolved photoluminescence (TRPL) measurements, we have used a Ti:sapphire femtosecond pulsed laser (the third harmonics $\lambda = 266$ nm) for excitation and a Hamamatsu synchroscan streak camera (time resolution of 20 ps) for detection of signal, respectively. The sample could be kept at different temperatures between 5 and 300 K using a liquid helium cooled cryostat. More details about dynamic measurements of the QW exciton in these structures can be found in Ref. [3].

Figure 1. Normalized PL spectra are shown for 10 and 50 K for the bare QW (solid blue lines) and for the hybrid with the cap layer thickness of 3 nm (dashed red lines). PL spectra for the same hybrid measured after 1 year are shown by black dotted lines. The hybrid structure is schematically drawn in the inset. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Results and discussion

PL spectra have been measured between 5 and 290 K for the bare QW structures (blue solid lines) and for the hybrids (red dashed lines) and are found to be rather similar for all samples. PL shows a dominant emission corresponding to the QW exciton and a weak peak from the buffer layer as illustrated in Fig. 1 for 10 and 50 K. In addition, the measurements are shown for the hybrid kept in atmospheric environment during 1 year (black dotted lines). These hybrids were rather stable within the weeks after preparation in difference from the hybrids fabricated with polymers. Nevertheless, here we also observe a degradation of the NRET process after a longer period. Dynamic properties of the QW excitons have been studied by temperature dependent TRPL. The PL decay curves are shown for 10, 50 and 290 K in Fig. 2 for the sample with the cap layer thickness of 3 nm with (red color) and without ZnO NCs film (blue color). Only for this sample the recombination rate increases in the hybrid, which was especially pronounced at temperatures of 30–100 K. The effect was opposite for the hybrid with 6 nm cap layer thickness, while in the control sample with 9 nm cap layer we have not observed any influence of ZnO NCs on the QW exciton dynamics. Possible reasons for such behavior are discussed in Ref. [3]. The QW exciton lifetime, τ , has been extracted by fitting the experimental PL decay curves using a single exponential decay function. This is a rather reasonable approximation though a small deviation from the law at temperatures QWs near the surface and the effect might be

opposite compared to NRET. The influence of the surface potential on luminescence properties in polar III-nitride heterostructures has been considered previously [7,8]. The TRPL measurements have been repeated for the hybrid structures after a period of 1 year to check if the samples fabricated using colloidal NCs are stable. We have found that even though using ZnO NCs, which are stable and have not changed their luminescence properties, the effect of NRET is vanished in the hybrid as shown in Fig. 2 (black lines) and Fig. 3 (black squares) for PL decay curves and the QW exciton lifetime, respectively. It is seen, that the PL decay is slower in the hybrid compared to the bare QW at all temperatures. The results suggest that the interface between the film and the QW structure might be affected with time due to humidity and/or due to thermal treatment such cooling–heating between cryogenic and room temperatures. Thus, the distance d can exceed 3 nm. In this case, the recombination rate in the QW layer will be affected mainly by the surface potential.

Figure 2. PL decay curves measured for the bare QW (blue lines), for the sample with ZnO NCs directly after coating (red lines) and after approximately 1 year. Results are shown for the structure with the cap layer thickness of 3 nm. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Figure 3. PL recombination time vs temperature extracted for the bare QW (triangles) and hybrid structure with 3 nm cap layer (circles). PL recombination time was also extracted for the same hybrid, measured after 1 year after preparation (squares).

Figure 4. NRET efficiency as a function of temperatures shown for the hybrid structure with 3 nm cap layer measured directly after fabrication.

Conclusion

The hybrid structures were fabricated to utilize NRET from the excitation generated in the AlGaIn/GaN QW to excitons in the colloidal ZnO NCs film. A difference between the PL recombination rate in the hybrid and in the bare QW structures becomes significant for the structure with 3 nm cap layer. The optimum pumping efficiency assuming the NRET recombination channel was found to be 42% at 60 K. No NRET effect has been detected in the control samples with 6 and 9 nm cap layer thickness, respectively. It is important to consider other factors affecting the exciton lifetime in QW in hybrids, for example, variation of the surface potential at the interface between ZnO and AlGaIn and also a degradation of the interface quality due to thermal treatment or as a result of ambient effect.

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