White light generating nonradiative energy transfer directly from epitaxial quantum wells to colloidal nanocrystal quantum dots

Sedat Nizamoglu[1], Emre Sari[1], Jong-Hyeob Baek[2], In-Hwan Lee[3] and Hilmi Volkan Demir[1]

[1]*Department of Electrical and Electronics Engineering, Department of Physics, Nanotechnology Research Center, and Institute of Materials Science and Nanotechnology, Bilkent University, Ankara, Turkey TR-06800*
[2]*Korea Photonics Technology Institute, Gwangju 500-460, Korea*
[3]*School of Advanced Materials Engineering, Research Center of Industrial Technology, Chonbuk National University, Chonju 561-756, Korea*
Email: volkan@bilkent.edu.tr

Abstract: We present white light generating nonradiative Förster resonance energy transfer at a rate of $(2ns)^{-1}$ directly from epitaxial InGaN/GaN quantum wells to CdSe/ZnS heteronanocrystals in their close proximity at chromaticity-coordinates (x,y)=(0.42,0.39) and correlated-color–temperature CCT=3135K.

OCIS codes: (230.5590) Quantum-well, -wire and -dot devices; (250.5590) Quantum-well, -wire and -dot devices

Solid state lighting (SSL) provides benefits in terms of significant reduction in electricity consumption and thus carbon dioxide emission [1]. As a result, SSL based white light generation has attracted significant attention worldwide. Today phosphors based color conversion approach is the most commonly used one. In this approach the blue electroluminescence of InGaN/GaN light emitting diode (LED) optically excites the phosphor, and the electroluminescence of the LED and photoluminescence of the phosphor together generate the white light spectrum. However, the limited blue absorption of phosphor requires using rather thick color converter, which leads to nonuniformity and undesired color change [1]. As an alternative to optical pumping, Förster resonance energy transfer (FRET) directly from semiconductor quantum wells (QWs) to color converters enables the use of relatively thinner color converter. To this end, in our study as the first proof-of-concept demonstration in solid state lighting we propose and demonstrate white light generating Förster resonance energy transfer pumping of colloidal semiconductor nanocrystal quantum dots by epitaxial quantum wells as shown in Fig. 1 [2].

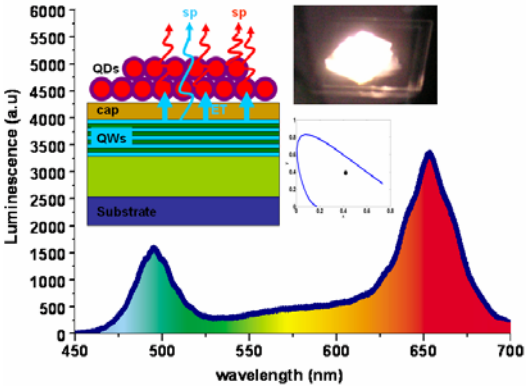
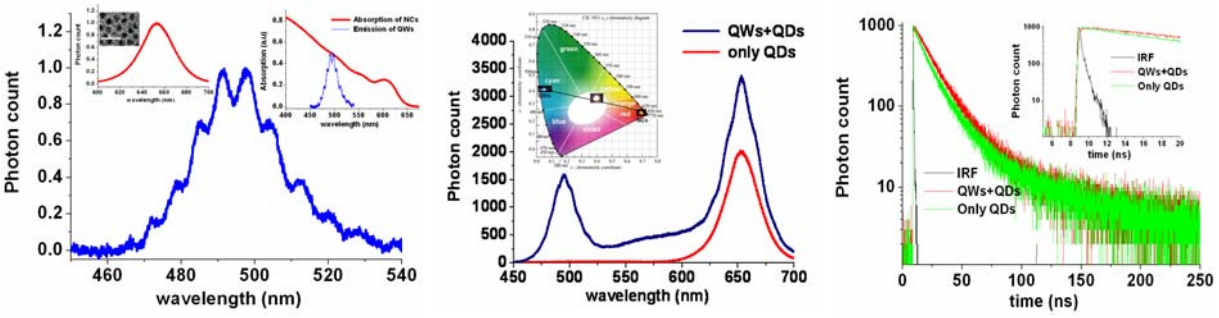


Fig. 1. Emission spectrum of our hybrid system improved by using Förster resonance energy transfer from cyan-emitting InGaN/GaN quantum wells (λ_{PL}= 490 nm) to red-emitting CdSe/ZnS core/shell nanocrystals (λ_{PL}= 650 nm), along with a device schematic of the white emitting hybrid system, its corresponding (x, y) chromaticity coordinates, and a photograph of the white emission.

To achieve white light generation we use cyan-emitting InGaN/GaN semiconductor quantum wells (QWs) (λ_{PL}= 490 nm) and red-emitting CdSe/ZnS core/shell nanocrystals (NCs) (λ_{PL}= 650 nm). For the InGaN/GaN epitaxial wafer, on top of the polished sapphire substrate we grow 3.5 μm n-doped GaN and subsequently 5 InGaN/GaN well/barrier epi-layers. Finally we finish our epi-structure with a few monolayers undoped GaN capping layer. The photoluminescence (PL) of our InGaN/GaN epi-wafer is shown in Fig. 2(a). The resulting optical emission peak of our epi-wafer is around 490 nm and its observed wavy PL is due to the phase separation in the quantum wells and barriers. For quantum dots, we use our NCs with diameters around 5.8 nm, which leads to PL peak around 650 nm. The spectral overlap of our NC absorption and QW emission make the dipole-dipole interaction possible between the transition dipoles of QWs and NCs as shown in the inset of Fig. 2(a).



(a) **(b)** **(c)**

Figure 2. (a) Photoluminescence of our epitaxial wafer around at 490 nm excited at 325 nm along with the optical absorption of NCs overlapping with the emission of QWs and also PL of NCs with their transmission electron microscopy image, (b) steady state emission spectrum of hybrid system (QWs+QDs) with respect to the only nanocrystal case (only QDs) along with the 1931 CIE chromaticity diagram showing the (x,y) operating point by using the combination of cyan-emitting quantum wells and red-emitting nanocrystals, and (c) time resolved spectroscopy measurement at 650 nm using a time resolution of 16 ps of the hybrid system consisting of quantum wells and nanocrystals together (QWs+QDs), compared to the only nanocrystal case (only QDs).

In Fig. 2(b), the steady-state photoluminescence of our hybrid system (i.e., nanocrystals integrated on quantum wells) is shown in comparison with that of the only nanocrystal case. The emission of the nanocrystals in the hybrid system is enhanced by 63% with respect to the only NCs case because of the energy transfer from quantum wells to nanocrystals. Furthermore, in the hybrid system the emission of nanocrystals exhibits a disturbed emission profile and this asymmetric luminescence is another indicator of FRET. It is a known fact that homogenous energy transfer also takes place among the nanocrystals due to their size distribution (<5%), however homogenous energy transfer would not give rise to such a wavy symmetry break in the PL spectrum. It is also worth discussing that in our hybrid structure FRET is a result of the dipole-dipole interaction rather than the delocalized behavior of excitons or free carriers, because the ZnS barrier of our nanocrystals prevent tunneling [3]. Here cyan-emitting quantum wells and red-emitting nanocrystals are chosen to make white light generation possible. For that using dual color emission the chromaticity line that connects both of the operating wavelengths of red NCs and cyan QWs should intersect with the white region as shown in the inset of Fig. 2(b), which makes it possible for our operating point to fall in white region if the color weights are arranged properly. In our implementation, the resultant white light corresponds to the chromaticity coordinates of (x,y)=(0.42, 0.39) and correlated color temperature CCT= 3135 K in the white region of CIE 1931 chromaticity diagram. Here FRET offers an effective balance in white light generation between the contribution colors, while also moving the operating point to a warmer color temperature. Figure 2(c) shows time resolved spectroscopy of the hybrid system and only nanocrystal solids. The emission increase for the hybrid structure reveals a smoother change from rise to decay around from 8 ns to 10 ns. This is due to the energy feeding component coming from FRET as also shown in the inset of Fig. 2(c). For only NCs on quartz substrate, we use double exponential fit with a $\chi 2$ of near unity ($\chi 2$=0.9369). The extracted lifetimes are 12.870 ns and 49.990 ns. The 12.870 decay component is the general lifetime of the nanocrystals, which is expected to be on the order of tens of nanoseconds. We attribute the other slow decay component (49.990 ns) to trap states. When we analyze the white light generating hybrid system QWs-NCs together, we already have double exponential lifetimes of 12.870 ns and 49.990 ns, which are the same with the only nanocrystal case. Furthermore, we find a third increasing exponential component of 2 ns, leading to an energy enhancement, which is the FRET component from quantum wells to nanocrystals for white light generation, with a $\chi 2$ of near unity ($\chi 2$=1.0171).

In conclusion, we proposed and demonstrated white light generating nonradiative FRET directly from epitaxial quantum wells to nanocrystal luminophors. Such white light generating FRET allows for the direct energy transfer to the color converting nanocrystals.

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