

Two-photon Induced Refractive Index Change In Quantum Dot Doped Photorefractive Polymer

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Abstract: Quantum-dot (QD) surfaces were engineered for two-photon (2P) induced localized photorefractivity. The use of sulfur rich surfaced QDs not only optimized charge transfer and resultant refractive index change but expanded the optical recording thresholds.

The key processes of the photorefractive effect are the conversion of the incident photons to separated free space charges and the consequent nonlinear effect for refractive index change. It was recognized that charge separation and transfer can occur at the interface of QDs and conjugated polymers with an electron affinity gradient even in the absence of an electric field. We have employed the successive ionic layer adsorption and reaction (SILAR) method to tune the surface stoichiometry of CdS QDs to optimize 2P induced charge transfer and consequent nonlinear response of dye molecules.

CdS core particles were prepared following a well established method [1]. Tuning the surface stoichiometry of CdS QDs was achieved by systematically injecting controlled amounts of additional S precursors [2]. The gradual deposition of unpassivated S atoms will significantly quench the radiative emission therefore enhancing charge transfer rate (see Fig. 1a).

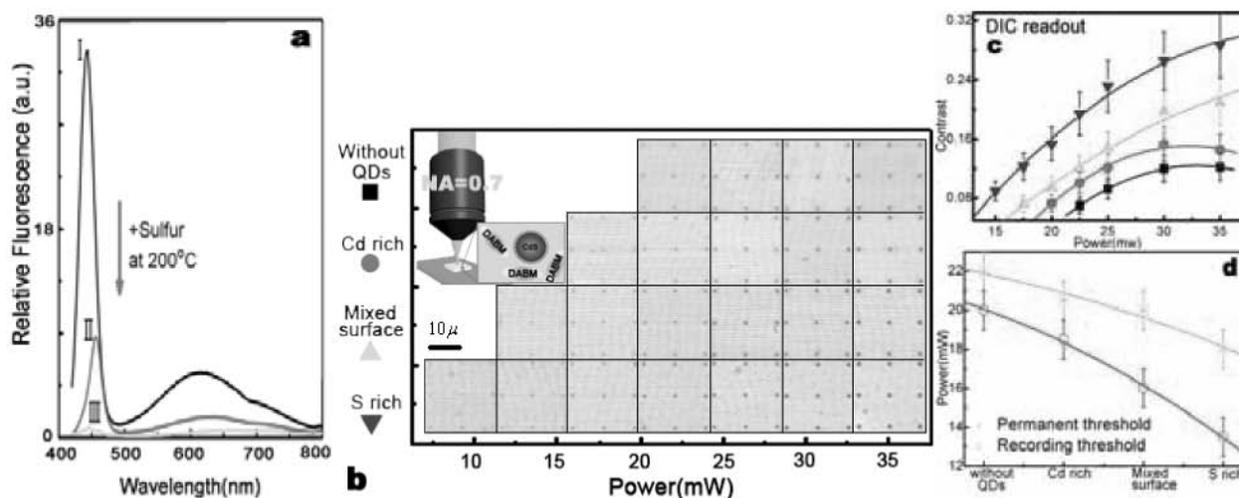


Fig.1 (a) dynamic evolution of fluorescence spectra. I, II and III correspond to QDs of the Cd rich surface, the mixed surface and the S rich surface respectively. (b) PL readout images of recorded bits in four different samples. (c) DIC readout as a function of recording power. (d) The recording threshold power (red circle) and permanent recording threshold power (green asterisk) as a function of surface modification.

Polymer samples consisting of poly(vinyl carbazole) (PVK), ethyl carbazole (ECZ), 4-diethylaminobenzylidene)-malononitrile (DABM) and QDs at the concentration ratio of 54mg:16mg:30mg:0.12nmol were prepared. A Ti:sapphire ultrashort pulsed laser (Spectra-Physics Tsunami) at wavelength 780 nm was employed for 2P excitation. Fig.1b shows the PL readout of recorded bits under 2P excitation. The fluorescence intensity of DABM is substantially depressed upon radiation where QDs were present, indicating the surface charge transfer between the DABM molecules and QDs. Furthermore, as the surface of the QDs was engineered from Cd rich to S rich the fluorescence was suppressed significantly although not completely. As a consequence of the space charge separation the local electric field change can introduce a change in refractive index through a possible electro-optic effect in DABM molecules which was substantiated by DIC readout, as shown in Fig. 1c. Interestingly, the margin between the threshold of erasable and permanent recording power is also enlarged as the gradual transition of the QD surface from Cd to S rich (see Fig. 1d).

References

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