Use of semiconductor nanocrystals for spectrally encoded high-density optical data storage

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Abstract: In this paper, we discuss the spectral encoding capability of semiconductor nanocrystals (NCs) and its application to high-density optical data storage. The effect of polymer matrix and energy transfer between the NCs are also studied.

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Three-dimensional (3-D) optical storage has received much attention for next generation optical storage technique due to its ability to overcome the current two-dimensional optical storage capacity limit imposed by the diffraction limit of light [1-3]. The heart of the three-dimensional storage technology is the two-photon absorption technique, in which two incident photons of a recording beam of an infrared wavelength are simultaneously absorbed by a recording medium to induce a local physical and chemical alteration. In previous efforts we saw successful developments of various types of two-photon-induced recording techniques, i.e., localised photorefractivity in photorefractive polymer [1], localised polarisation sensitivity in a polymer-dispersed liquid crystal (PDLC) material [2] for rewritable (or erasable) 3-D bit optical storage, and localised void formation in photorefractive polymers [3].

More recently, we have demonstrated the 3-D recording on the densely packed CdSe/ZnSe/ZnS semiconductor nanocrystal thin film, where two-photon absorbed luminescence enhancement was employed as the recording mechanism [4]. The significance of this work is the fact that the spectral properties of NC such as emission wavelength tuneability and narrow emission bandwidths can further be incorporated to produce spectrally encoded high-density optical storage. In this paper, we present the recent progress of study on spectral encoding capability of semiconductor nanocrystals (NCs) and its application to high-density optical data storage. In particular, the effect of polymer host matrix and the energy transfer between the NCs are also studied.

In order to demonstrate the spectral encoding capability of NCs, a densely packed thin film of two size NCs is prepared. The NCs and the film are synthesized and prepared as in Ref. 4. The two sizes of NCs (2.5nm and 6nm core) had emissions at 550nm and 620nm respectively. Figure 1 shows the emission spectrum of the thin film, excited by Ti:Sa femtosecond pulsed light at 800nm (80MHz).

![Fig. 1. Emission spectrum of two sizes of NCs in a densely packed thin film. The two core sizes of NCs are 2.5nm and 6nm and had emissions at 550nm and 620nm respectively.](image)

In Figure 2, the data recording by two-photon absorption at various wavelengths are shown, where the spectrally resolved images of the recorded pattern is shown. Notice that at wavelength 980nm (letter R), the reading image at 620nm shows clear emission enhancement at the exposed region, whereas the reading image at 550nm shows negligible enhancement.
Currently, there are several possible mechanisms that can be responsible for the observed spectral encoding on NCs. Firstly it could be possible by the fact that the two-photon absorption cross-section spectra (with respect to absorption wavelengths) of two sizes of NC are necessarily different [5, 6]. This means that at certain wavelengths, certain size of NCs show relatively improved absorption than other NCs, thus causing the difference in corresponding photo-enhancement speeds so that marks can be recorded on one type of NCs without affecting too much of the other NCs. In such scenario, we should be able to record each colour separately. However, our experimental results showed that only the recording on red NCs was possible.

Secondly, the electronic energy transfer between the two sizes of NCs [7] could cause the change in enhancement speed. Since the emission of the green NC overlap with the absorption of red NCs, the enhancement of red NCs could be facilitated at the expense of slowing green enhancement. In this case, we would only be able to record on red NCs. This is a plausible explanation for what we observe in Figure 2. However, we expect the two proposed mechanisms be affecting the results simultaneously.

Further investigation into the energy transfer induced photo-enhancement was conducted and the results are shown in Table 1. Since the energy transfer is dependent on the neighbouring NC distances, we homogeneously doped the NCs in polymethylmethacrylate (PMMA) matrix to separate the distance between NCs and observed their enhancement kinetics. The enhancement was fitted to the power-law ($y = ax^b$) and the power index $b$ was recorded [4].

<table>
<thead>
<tr>
<th>Sample</th>
<th>Distance between surfaces</th>
<th>Red NCs only (unmixed)</th>
<th>Red mixed with green</th>
<th>Green NCs only (unmixed)</th>
<th>Green mixed with red</th>
</tr>
</thead>
<tbody>
<tr>
<td>Densely packed thin film</td>
<td>~ 2 nm</td>
<td>0.11</td>
<td>0.2</td>
<td>0.4</td>
<td>0.24</td>
</tr>
<tr>
<td>Ultra-high concentration in PMMA</td>
<td>~ 4 nm</td>
<td>0.15</td>
<td>0.34</td>
<td>0.28</td>
<td>0.25</td>
</tr>
<tr>
<td>High concentration in PMMA</td>
<td>~ 7 nm</td>
<td>0.23</td>
<td>0.3</td>
<td>0.18</td>
<td>0.25</td>
</tr>
<tr>
<td>Low concentration in PMMA</td>
<td>&gt; 20 nm</td>
<td>0.48</td>
<td>0.43</td>
<td>0.27</td>
<td>0.29</td>
</tr>
</tbody>
</table>

Note that for high concentrations (i.e., distance between NCs less than 7nm, densely packed and ultra-high concentration), the red NCs tends to photo-enhance faster in the mixed sample than in the unmixed sample, while green gets slower in the mixed. This situation is not valid anymore for low concentration cases, where the distance between NC surfaces gets larger than 20nm and the index values are similar. This clearly shows that the energy transfer plays the major role in the observed spectral encoding. Further results on spectral encoding capabilities of NCs shall be presented.
References