單一硒化鎘/硫化鋅膠體量子點之螢光特性

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中文摘要

本論文探討由化學合成的單顆硒化鎘/硫化鋅膠體量子點之光學特性。單量子點的螢光閃爍，是由歐傑過程、熱過程、或穿隧效應所引起的量子點離子化，使單量子點的發光行為，會隨著時間的進行而呈現明暗交替。為了研究螢光的閃爍行為，首先我們改變激發光源的強度。再將量子點所處的周遭環境，由絕緣體更換為導體。最後改變量子點的大小。此外，我們在量子點的表面接上有機鏈，觀察其光性的變化。我們的量測除了主要針對單顆量子點進行時間解析實驗外，並且比較量子點群的光激螢光和吸收光譜。對於改變激發強度的部份，螢光閃爍的發光持續時間和閃爍的頻率隨著激發光強度呈現急劇變化，然而不發光的持續時間和激發強度無關。另外，當激發光強度增加時，輻射的生命週期呈現減弱的趨勢。當量子點所處的環境由絕緣體換成導體(混入金奈米粒子)時，發現螢光閃爍的閃爍頻率被抑

止、發光持續時間延長、且同時螢光的強度有明顯的增加。有關改變量子點大小的部份，並沒有觀察到螢光閃爍隨量子點大小改變的趨勢。

另外，我們在硒化鎘和硒化鎘/硫化鋅量子點表面接上 TOPO(三辛基磷化氫氧化物),HDA(正十六碳胺),和 PA（pylamine）有機分子鏈。從整體的螢光光譜來看，對於硒化鎘量子點而言，我們發現接上 TOPO-HDA 後，螢光強度為最小；對硒化鎘/硫化鋅而言，卻為最大。在單顆的量子點的情況，相較之下，我們發現對於螢光閃爍 TOPO-HDA-PA 有較長的發光持續時間。另外，我們利用 Kohlrausch-William-Watts 公式來決定生命週期分佈的程度。我們發現由接上 TOPO-HDA 的量子點，生命週期對於硒化鎘有最大的分佈，對硒化鎘/硫化鋅卻為最小。當 TOPO-HDA 接合上後，再加入 PA 有機分子鏈，對於硒化鎘和硒化鎘/硫化鋅將會減少生命週期的分佈。而比較量子點群和單量子點，其螢光強度的對應為相符的。

Fluorescence properties of single CdSe/ZnS colloidal quantum dots

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ABSTRACT

In this thesis, we study the optical properties of single CdSe/ZnS colloidal quantum dot. One of the interesting phenomenons of single quantum dot is the fluorescence intermittency. Due to the ionization of quantum dot by Auger, thermal, or tunneling process, the bright and dark phenomena alternatively exist. The effects of excitation power and surrounding matrix on the blinking behavior were studied. We also investigated the influences of quantum dot size and organic ligand conjugation on the optical properties of quantum dot. For excitation power dependence study, it shows that the emissive time interval and the blinking frequency of fluorescence intermittency dramatically changes by excitation power, however, the non-emissive time interval is independent of excitation power. In addition, the radiative lifetime decreases while the excitation power was increased. Moreover, the fluorescence

intermittence behavior was suppressed when the surrounding matrix was changed from insulation glass cover-slip to conducting gold nano-particles, and also the fluorescence intensity could be enhanced by the plasmon resonance. For quantum dot size dependent investigation, no correlation was observed between quantum dot size and fluorescence intermittency.

In addition, the TOPO (trioctylphosphine oxide), HDA (hexadecylamine), and PA (pylamine) organic ligands were conjugated to the CdSe/ZnS and CdSe quantum dot surface. For QD clusters, the TOPO-HDA conjugated quantum dot has the lowest fluorescence intensity than the others. On the contrary, it is the highest in CdSe/ZnS QD. On the other hand, for the single QD, the fluorescence intermittency for the TOPO-HDA-PA ligand conjugated QD has larger emissive/non-emissive ratio. Besides, we use Kohlrausch-William-Watts stretching exponential function for radiative decay curve fitting to determine the degree of lifetime distribution. We found that the TOPO-HDA ligand conjugated QD has largest lifetime distribution in bare CdSe case, but smallest one for CdSe/ZnS. After TOPO-HDA ligand conjugated, adding the PA ligand would reduce lifetime distribution for both CdSe and CdSe/ZnS quantum dot. The comparison of integrated fluorescence of single quantum dots is corresponding to that of ensemble average.