Kwansei Gakuin University
Report of Research Outcome

2023/03/13

To President

Department : Science and Technology
Position : Postdoctoral fellow
Name : Li Wang

I report the outcome of the research as follows.

Name of the Fund/Program	□Sabbatical leave with grant □Sabbatical leave with no grant □KGU Joint Research □Individual Special Research ☒Postdoctoral fellow ※Please report by designated form as for “International Research Collaboration”.
Research Theme	Elementary carrier dynamics of blue-emissive semiconductor quantum dots controlled by interfacial engineering
Research Site/Venue	Tamai Lab.
Research period	2022/04/01 ～ 2023/03/31 （12 month）

◆ **Summary of the research outcome** （**approx. 2,500 words**）

Please write down the outcomes in detail regarding the research theme above.

Colloidal quantum dots (QDs) exhibit unique size-tunable optoelectronic properties due to strong quantum confinement effects, making them as potential candidates for optoelectronic devices. For applications such as lasers, displays, and nanosensors that require high transition strength with a narrow bandwidth, the spectroscopy and dynamics of QDs are determined by size-distribution, exciton-phonon coupling, and thermal active energy. These factors can be studied through temperature-dependent photoluminescence (PL) experiments. Recently, there has been growing interest in heavy metal-free QDs as a means to reduce their environmental impact. ZnSe with 2.7 eV bulk bandgap is a suitable choice for violet-blue nano-emitters. In this study, ZnSe QDs (426 nm) prepared by a hot-injection method were dispersed in a poly methyl methacrylate (PMMA) matrix or drop-casted on a quartz plate for temperature-dependence experiments. PL spectra of QD films were measured in a Montana cryostation (from 295 to 5 K) with ~400 mW/cm^2 365 nm CW or UV fs excitation, and detected in a transmission mode by a Princeton Spec-10 CCD. Moreover, ZnSe/ZnS core/shell (C/S) QDs were dissolved in isopentane (OD_{300nm} = 0.15, core QD 397 nm). Temperature dependent PL of C/S QDs were measured by Fluorolog-3, and temperature was controlled by UnispeKs (>80 K).

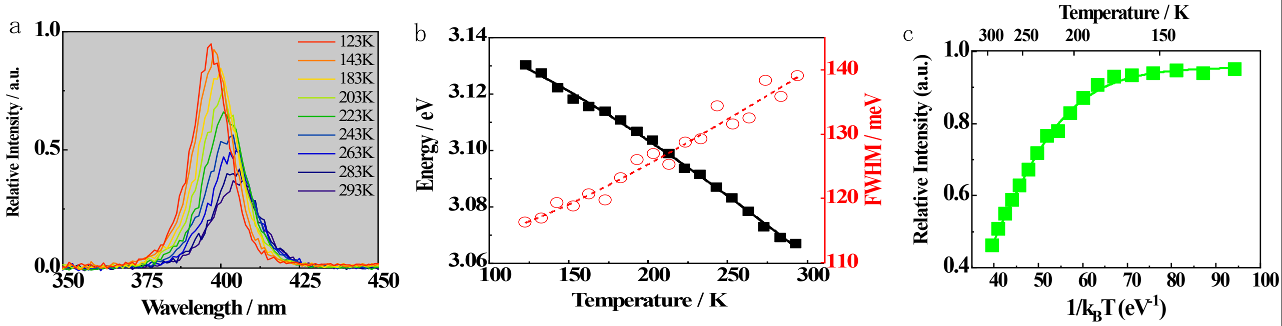

Figure 1. (a) Temperature dependent PL spectra, (b) PL peak energy and FWHM, and (c) PL intensity of ZnSe/ZnS core/shell QDs with PL peak at 406 nm in isopentane.

ZnSe/ZnS C/S QDs with 1S peak at 397 nm were washed with hexane/acetone mixture (1:4),and then the deposition was dissolved in isopentane (QY, 18%). Temperature-dependent PL spectra, peak energy, and the full width at half-maximum (FWHM) of C/S QDs in isopentane are shown in Figure 1. As the temperature increases, the PL intensity decreases along with a red-shifted emission energy and a broadened FWHM. The redshift is well fitted to the Varshni relation $E_g(T) = E_g(0) - \alpha T^2/(T+\beta)$, where $E_g(0)$ is the energy gap at 0 K, α is the temperature coefficient, and β has a value close to the Debye temperature of the material.[1] The broadening of the FWHM can be well explained by exciton coupling with acoustic and LO phonons, which can be described by the following equation $\Gamma(T) = \Gamma_{inh} + \sigma T + \Gamma_{LO}/(e^{E_{LO}/k_BT-1})$, where Γ_{inh} represents the temperature-independent inhomogeneous distribution resulting from the size fluctuations. σ is the exciton-acoustic phonon coupling coefficient and Γ_{LO} represents the strength of exciton-LO phonon coupling. E_{LO} is the LO-phonon energy and k_B is the Boltzmann constant. The temperature-dependent PL intensity was analyzed to reveal the nonradiative processes. The integrated PL intensity was plotted as a function of $1/k_BT$, as shown in Fig. 1(c). It is evident that the PL intensity remains almost constant up to 170 K and then rapidly decreases. Considering the thermally activated carrier escape (with activation energy E_a), the temperature dependence of PL intensity can be described by $I_{PL}(T) = I_0/(1 + ae^{(-E_a/(k_B T))})$,where $I_{PL}(T)$ is the integrated PL intensity at temperature T and I_0 is the integrated PL intensity at 0 K. The experiment data are well fitted with the constants listed in Table 1. The activation energy E_a corresponds to the exciton binding energy. For small sizes, the binding energy is larger. The results will be further discussed in terms of quantum confinement effects.

		Core426	C/S397
$E_g(0)$	eV	2.74	3.15
α	meV/K	0.11	0.61
β	K	340	332
Γ_{inh}	meV	105	129
σ	μeV/K	7.6	18
Γ_{LO}	meV	17	53
E_{LO}	meV	20	39
E_a	meV	33	103

Reference:

[1] Valerini *et al.*, Anni, *Phys. Rev. B* **2005**, *71*, 235409.

Conference:

1. R. Takeuchi, D. Eguchi, N. Tamai, L. Wang, "Localized Surface Plasmon Resonance in Gold Nanorods Under Ultrahigh Pressure", The 103rd CSJ Annual Meeting, K205-2vn-03

Publication:

1. S. Jung, L. Wang, H. Sugiyama, H. Uekusa, T. Katayama, K. Kamada, T. Hamura, and N. Tamai, "Intramolecular Singlet Fission in Pentacene Oligomers via an Intermediate State" (J.

Deadline：Within two months after finishing the research period.

Sabbatical leave with grant: Submit this report to President with confirmation by the dean of school you belong to.

※Postdoctoral fellow is required to submit this report with confirmation by the dean of graduate school before the end of employment period.

Where to submit：Organization for Research and Development and Outreach (NUC)

◆We put this report on the web of KGU. If there is any problem about it because of difficulties on your research, please let us know.