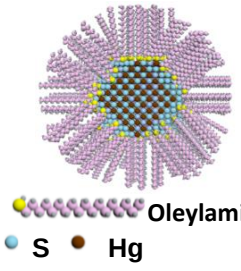
The electronic dynamics of conduction band of OLA-HgS Quantum dot

Joonhyung Lim[1,2], Bitna Yoon[2], Kwang Seob Jeong[2], Kyungwon Kwak[1,2], Minhaeng Cho[1,2]

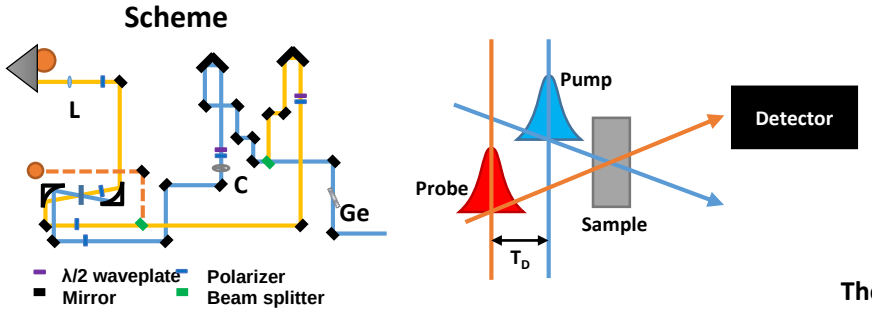
1. Center for Molecular Spectroscopy and Dynamics, Institute for Basic Science (IBS), Seoul 02841, Republic of Korea,
2. Department of Chemistry, Korea University, Seoul 02841, Republic of Korea, Email:kmno0831@korea.ac.kr

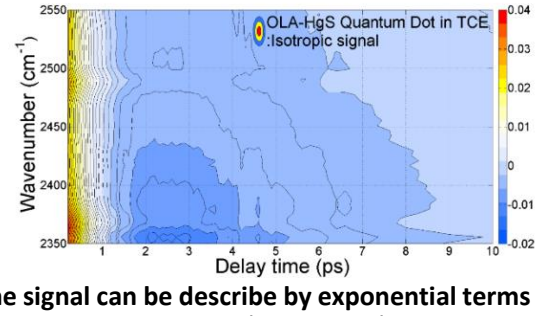
Introduction



The HgS Collidal Quantum Dots(CQDs) with Mid-IR intraband gap can be fascinating materials for IR photodevices[a]. Here, we reported the interesting relaxation dynamics of Oleylamine-doped HgS CQDs which have size-dependent intraband gap[b]. By using IR pump-probe techniques, we observed frequency-independent fast decaying dynamics (1.2 ± 0.1ps) accelerated by Auger process in CQDs with biexciton generation and decay process (>300 ps) caused by Phonon bottleneck for CQDs with single photon absorption. Moreover, the dip(~2ps) is observed, and assigned to the red-shift from the carrier-carrier interaction in this measurement.

The scheme of Pump-Probe setup & Analysis

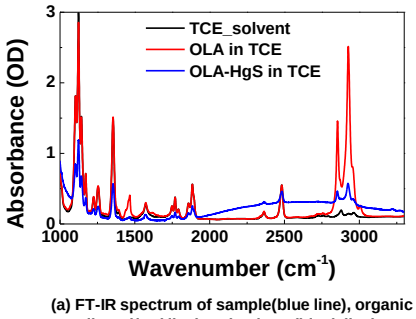




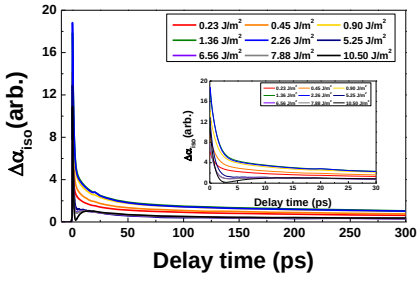
The signal can be describe by exponential terms

$$\left(\Delta\alpha_{iso} = A_1 \cdot e^{-\frac{t}{\tau_1}} + A_2 \cdot e^{-\frac{t}{\tau_2}} + \cdots\right)$$

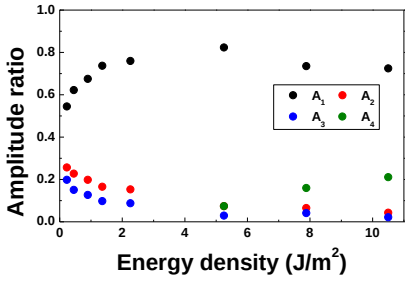
Experimental Results



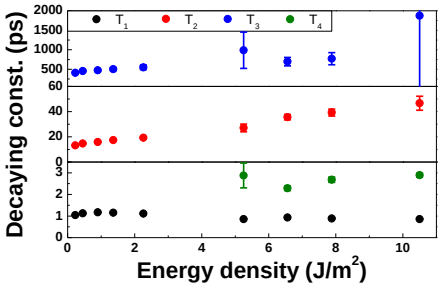
(a) FT-IR spectrum of sample(blue line), organic ligand(red line) and solvent(black line)



(b) The isotropic T.A. with various energy density of pump pulse(0.25~10.50 J/m2), the inset box describe the dip

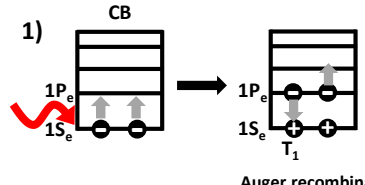


(c) The amplitude ratio of each components used to describe the decaying pattern fitting functions descriptions



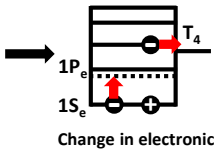
(d) The decaying constants of each components to describe the electronic dynamics

Physical mechanism

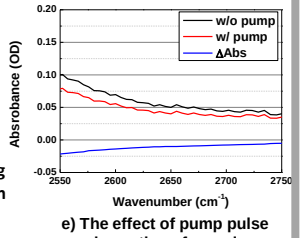


Biexciton generation with strong pump pulse

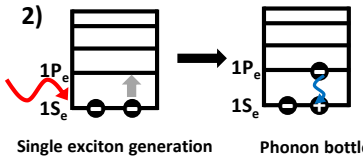
Auger recombination (energy transfer to another carrier)[c]



Change in electronic structure from binding energy[d] and relaxation scheme_phillippe



e) The effect of pump pulse on absorption of sample



Single exciton generation with weak pump pulse

Phonon bottleneck[5] (very slow relaxation, >300ps)

Discussion & References

With the pump-power dependency and decaying constants of each components, we can successively label the each components. Especially, the components 4 (slow components) makes this quantum dot as the candidates for photodevices.

(a) Lhuillier, E.; Scarafagio, M.; Hease, P.; Nadal, B.; Aubin, H.; Xu, X. Z.; Lequeux, N.; Patriarche, G.; Ithurria, S.; Dubertret, B., Infrared Photodetection Based on Colloidal Quantum-Dot Films with High Mobility and Optical Absorption up to THz. *Nano Lett* **2016,** *16* (2), 1282-6.
(b) Yoon, B.; Jeong, J.; Jeong, K. S., Higher Quantum State Transitions in Colloidal Quantum Dot with Heavy Electron Doping. *J Phys Chem C* **2016,** *120* (38), 22062-22068.
(c) Rabouw, F. T.; Vaxenburg, R.; Bakulin, A. A.; van Dijk-Moes, R. J.; Bakker, H. J.; Rodina, A.; Lifshitz, E.; A, L. E.; Koenderink, A. F.; Vanmaekelbergh, D., Dynamics of Intraband and Interband Auger Processes in Colloidal Core-Shell Quantum Dots. *ACS Nano* **2015,** *9* (10), 10366-76.
(d) Klimov, V. V.; Hunsche, S.; Kurz, H., Biexciton effects in femtosecond nonlinear transmission of semiconductor quantum dots. *Phys Rev B Condens Matter* **1994,** *50* (11), 8110-8113.

Acknowledgement

This work was supported by the Institute for Basic Science (IBS) R023-D1