Quantum dots under high pressure: optical properties and x-ray diffraction analysis

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Introduction

Nanocrystal quantum dots (NQDs) are of great interest because of their unique physical properties and possible technological applications. Quantum dots can exhibit properties that are absent in bulk material [1]. Application of pressure can affect optical and other properties significantly. The pressure effects on the properties of PbSe are, however, known for the bulk material only.

Methods and Materials

We present studies of optical properties and crystal structure of PbSe quantum dots under pressure by FTIR and x-ray diffraction spectroscopy. Three NQDs sizes, 3, 5, and 7 nm in diameter, were examined. We measured absorption of PbSe NQDs in the range of pressures from ambient to more than 4 GPa. The x-ray diffraction data was collected at Advanced Photon Source in Argonne National Laboratory. The sample has been loaded into diamond-anvil cell (DAC) and exposed to the intense x-ray beam, with x-ray wavelength $\lambda = 0.368$ Å.

Results

Example of absorption spectra is shown in Fig. 1. We observe a red shift of the lowest-energy 1S exciton feature with increasing pressure. This is consistent with bulk PbSe. In Fig. 2, we present the pressure dependence of IR absorption for PbSe NQDs. We have also included the calculated effect from quantum confinement on absorption energy.

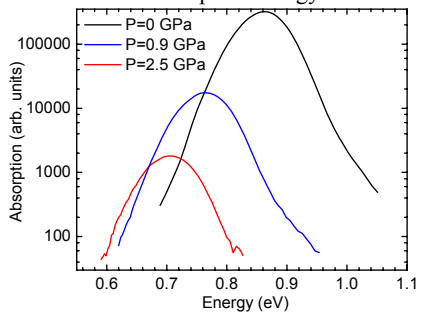

Fig. 1. Infrared absorption spectra of 5-nm diameter PbSe NQDs for three indicated pressures show a red shift of the lowest-energy 1S excitonic features with pressure.

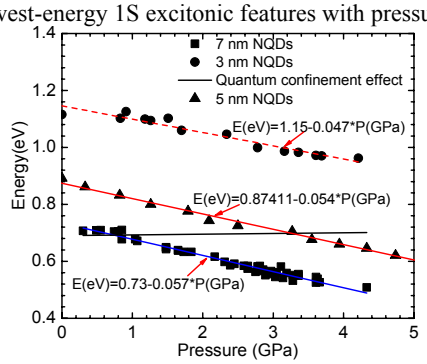

Fig. 2. Peak position of the 1S absorption feature as a function of pressure.

Preliminary data on x-ray spectra obtained at two different pressures are presented in Fig. 3. In Fig. 4 we present the pressure-volume data for 5 nm PbSe NQDs. The experimental data was fit to Murnaghan equation of state (EOS), with bulk modulus derivative B_0' fixed to 4.5.

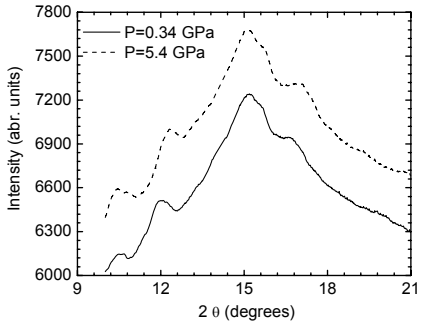

Fig. 3. X-ray spectra of PbSe NQDs at different pressures.

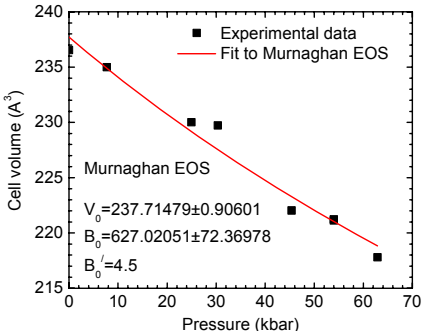

Fig. 4. Unit cell volume (in Å^3) as a function of pressure.

Discussion

For all sizes a large shift of absorption with pressure, 57 meV/GPa, 54 meV/GPa, and 47 meV/GPa for 7, 5, and 3 nm size NQDs, respectively, is observed. The observed effect is consistent with the shift that is expected to occur as a result of the deformation potential in the bulk material, which suggests that NQDs show almost no change due to volume reduction and quantum-confinement related effects due to the applied pressure. The lattice constant and bulk modulus for NQDs are slightly higher than those of the bulk material. It is known that phase transition in bulk PbSe occurs at about 4 GPa. Therefore the absence of phase transitions in our experiments can be attributed to the fact that it shifted to the pressures higher than 60 kbar.

Acknowledgements

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References:

[1]. Pietryga, J. M.; Schaller, R. D.; Werder, D.; Stewart, M. H.; Klimov, V. I.; Hollingsworth, J. A., *J. Am. Chem. Soc.*, **126**(38), 11752-3 (2004).