

Calculations of Size-Dependent Trions and Biexcitons Binding Energies in Lead Halide Perovskite Nanocrystals

T. Sato¹, K. Cho², H. Suzuura¹, and Y. Kanemitsu²

¹Graduate School of Engineering, Hokkaido University, Sapporo, 060-8628, Japan

²Institute for Chemical Research, Kyoto University, Gokasho Uji-city, Kyoto 611-0011, Japan

xs4qt6@eis.hokudai.ac.jp

We theoretically study the size dependence of the trion and biexciton binding energies of lead halide perovskite nanocrystals (NCs). Lead halide perovskite NCs are promising materials for optical devices due to their excellent properties such as band gap tunability due to the possibility of halogen substitution and high photoluminescence quantum yield. The exciton emission spectra of lead halide perovskite NCs obtained by single-dot spectroscopy under low-temperature conditions accompany the sidebands originating from multiexciton states, that is, trions and biexcitons [1], deeply involved in the application of NC-based devices, as they can affect the optical gain of lasers and reduce the luminescence efficiency of LEDs. Moreover, biexcitons can play an important role in the generation of entangled photon pairs. This is the reason why such exciton complexes in NCs attract much attention.

For the single exciton state in NCs, there is a good agreement between experiment and theory [2], but this is not the case with exciton complexes due to the fact that quite a large number of bases are necessary to accurately calculate their binding energies. Recently, there have been several theoretical studies, while the calculated binding energies underestimate the experimental results [3,4]. In this paper, we have performed theoretical calculations based on the effective mass model in a cubic geometry, which is close to the actual crystal structure, in order to quantitatively explain the emission spectrum of lead halide perovskite NCs.

First, we incorporated the dielectric confinement effect, which has been pointed out in previous studies as the cause of the underestimations for the binding energies of the exciton complexes. Certainly, this effect increases the binding energy. But that is not enough for explaining the experimental data theoretically [5].

Next, we considered the influence of the polaron effect. Since lead halide perovskite is an ionic crystal, lattice distortions can give rise to local deformation of dielectric constant that affects the binding energy of the exciton complex. We have found that what resolves the underestimations is to correctly evaluate the polaron effect for the bulk exciton. In fact, we have obtained the calculated results giving better fitting to the experimental data, using realistic values of optical/static dielectric constant and polaron radius by previous theoretical studies (Fig. 1).

This is how we can theoretically explain the binding energy of exciton complexes in lead halide perovskite NCs to a large extent by combining the dielectric confinement and polaron effects. In addition, we are going to discuss the effects of nonparabolicity in one-particle dispersion relations.

References

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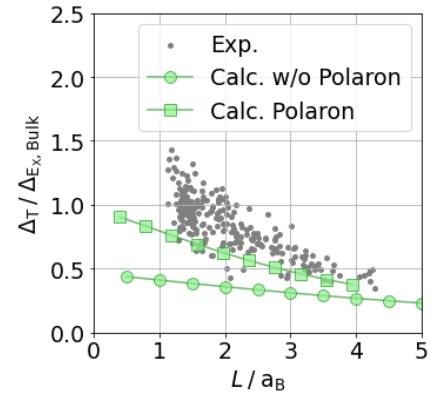


Fig.1. The size dependence of trion binding energy. Gray dots are experimental data, square and circle plots are calculated results with out or with polaron effect.