20th Brazilian Workshop on Semiconductor Physics

Controlling the Nonlinear Optical Response of Semiconductor Nanomaterials by Heterostructure Engineering

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Abstract

The possibility of controlling the optical and electronic structure in semiconductor by quantum confinement has turn semiconductor colloidal nanocrystals (NCs) one of the most investigates class of materials for optoelectronics applications. In the last two decades, much effort has been devoted to gain further control over those properties by electronic wavefunction engineering via core/shell heterostructuring. Despite all the advances in this field, very little is known about the influence of these heterostructures on the nonlinear optical response of NCs. Particularly, two-photon absorption, in the so-called quasi-continuum spectral range, has been shown to be linearly dependent on the NCs volume, on a quasi-universal scaling rule. [1] As a result, the most common method to increase the nonlinear absorption response in NCs is to make the nanomaterial larger, resulting on a red shift of their size-dependent bandgap. Here, we discuss how bandgap engineering by core/shell heterostructuring can be used to enhance the nonlinear optical response of NCs without drastically changing the emission energy. It will be shown that by wisely choosing the core and shell dimensions one can enhance the two-photon absorption cross section by one order of magnitude while maintaining the same emission energy. An alternative approach to further control the nonlinear optical response is to explore non-spherical shell as it is the case for dot-in-rod heterostructures. [2] With this type of NCs one can explore extra degrees of freedom in order to control the electronic properties and, ultimately, the nonlinear optical response.

References

[1] G. Nagamine et.al., J. Phys. Chem. Lett. 9(12), 3478 (2018)
[2] D. Kim, ACS Nano 11 (12), 12461 (2017)