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Spectrally Resolved Nonlinear Optical Properties of Colloidal Quantum Confined Semiconductor Dots, Rods and Nanoplatelets

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Extended Abstract

Semiconductor nanomaterials with dimensions close to the exciton Bohr radius have received great attention over recent years, owing to their unique physical and chemical properties [1]. Luminescent quantum dots (QDs) are ideal luminophores for multiplexed optical coding because their fluorescence emission band position can be continuously tuned by changing the particle size. The absorption and emission spectra are regarded as properties of the material and are normally independent of the light intensity. However, for sufficiently large light intensities, such as those available with femtosecond lasers, these optical characteristics additionally become functions of the light intensity. The quantum confinement and dielectric confinement effects make these materials a promising class of third-order nonlinear optical (NLO) media with large third-order nonlinear susceptibilities and a fast response time. The colloidal syntheses presented here allow the fabrication of morphology well-defined, monodisperse in size semiconductor QDs, QRs, or quantum nanoplatelets (NPLs). In the literature current studies are primarily the NLO properties measured usually at a single wavelength. We studied the nonlinear optical (NLO) properties of several quantum-confined semiconductors with various morphologies, i.e. quantum QDs, QRs and NPLs. The NLO properties of the samples were measured by the Z-scan and two-photon excited emission (TPEE) techniques using a femtosecond laser system in a wide spectra range. Here, we show the NLO factors in a wide wavelength range to find maximal values of the parameters and optimizing them. Thus, in this study, we provide both: fundamental knowledge about materials engineering of a new class of inorganic colloidal nanomaterials, i.e. optimization of synthesis and surface modification, and fundamental knowledge how to correctly characterize those materials using femtosecond laser techniques. Even more important, the colloidal synthesis presented in this work allows for the fabrication of well-defined monodisperse in size semiconductor QDs, QRs, and NPLs. For example, the nonlinear absorption of the spherical CdSe and CdS QDs was found to be the strongest close to twice the wavelength of the second exciton absorption band of the QDs with a maximum of 66,000 GM and 7,200 GM; respectively [2,3]. In one-dimensional CdSe QRs we observed significantly enhanced nonlinear response, where the two-photon absorption cross section σ_2 was found to be as large as 164,000 GM [4]. Two-dimensional CdSe NPLs of different thicknesses were synthesised and their wide range nonlinear optical properties were investigated, leading to determination of maximum values of two-photon absorption cross sections. 5.5 ML CdSe NPLs with two-photon absorption cross section of 8.0×10^4 GM were chosen as a model material for functionalization toward biological applications, especially in bioimaging [5]. Encapsulation in polymeric NCs was selected as a hydrophilization method, which also provided cell viability after incubation without any significant change in optical spectra and preserving nonlinear absorption and two-photon excited emission properties as well as being visible through a two-photon microscope. The two-photon cross section of polymeric NCs filled with 5.5 ML CdSe NPLs was estimated as 2.0×10^8 GM. The 4.5 ML CdSe NPLs were chosen for studies on the relation between noble metal doping and nonlinear optical properties. Doping with silver and copper ions led to two-photon absorption cross sections of up to 5.44×10^6 GM and 1.33×10^7 GM, respectively, and thus appears to be a method for enhancement of nonlinear optical properties of semiconductor NPLs without increasing their volume [6]. The features of these semiconductor optically active nanosystems relevant for multi-photon fluorescence microscopy applications will be briefly discussed [7].

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