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Short-range vibroelectronic coupling between quantum dots and organic molecules

Within colloidal quantum dots (QDs) research, the influence of the molecular surroundings on the optoelectronic properties of CdTeS semiconductor nanocrystals is intensively studied. In this context, our work consists in probing this QD/molecule coupling from the converse point of view: we examined the influence of the optoelectronic properties of QDs on their molecular environment and evidenced a correlation between the electronic structure of QDs and the vibrational structure of molecules. Thanks to Two-Color Sum-Frequency Generation nonlinear optical spectroscopy, combining visible and infrared laser beams on ligand-conjugated QDs, we demonstrate that the vibrational response of ligands is maximum when QDs are pumped into their excitonic states. Our formal calculations showed that a classical dipolar interaction can account for the results, considering that the polarizability of the molecules is subordinate to the local electric field induced by the excitons. In a similar way than Forster resonant energy transfers (FRET) which occur between two fluorophores, the dipole-dipole coupling between QDs and molecules proves to be short-range, and can thus be used for chemical sensing. But unlike FRET, this surface-specific coupling does not require any spectral overlap between the two interacting systems and allows the detection of any molecule without fluorescent label. This innovative result constitutes quite a good news for physical chemists and chemical physicists.

Choix de session parallèle

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