Fullerene-cluster amplifiers and nanophotonics of fullerene solutions

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Nanophotonics of fullerenes is mainly associated with the impact of low doping of fullerenes on characteristics of nonlinear optical (NLO) media and, as a result, of NLO devices. Obvious electromagnetic nature of NLO effects enhancement and a direct influence of electric field on the enhancement witness convincingly the recruitment of charge states in the considered features. These states have been explained by the formation of charge transfer complexes due to donor-acceptor (DA) interaction between fullerenes and host matrices. The paper concerns the origin, structure, and spatial extension of the complexes. Empirically, the complexes are studied via enhancement effects which follow Raman scattering and one-photon luminescence of fullerene-doped matrices. Computationally, the DA interaction is considered through its influence on the total intermolecular interaction between solute (fullerene) molecules (sol-sol interaction) as well as between solute and solvent molecules (sol-solv interaction).

Empirical features are exhibited by an additional blue emission spectrum in diluted fullerene-doped matrices which appears under excitation in the visible and near UV regions subjected to resonance conditions [1-3]. The blue spectrum presents a combination of enhanced Raman scattering and enhanced one-photon luminescence of both solvent and fullerene molecules, the latter involved in clusters, and is attributed to the enhancement of linear optical effects caused by the formation of sol-sol and sol-solv clusters. A particular contribution of these spectral components into the blue emission of a certain solution depends on chemical structure of the fullerene. Quantum-chemical computations performed in the study permitted to evaluate the strength of the intermolecular interaction in the clusters and highlight their charge-transfer origin.

The observed enhancement of linear optical events is considered within the framework of the enhancement local field model [4] and is connected with the resonance excitation of the clusters which are spatially extended charge-transfer complexes. The excitation light is transformed into local charge-transfer excitons within the clusters providing the polarization of the medium enough for the enhancement of the local electric field of the incident and outgoing light. A direct connection is established of the intensity of the observed linear optical features with the efficacy of the media nonlinear behavior, at one hand, and with a coupling strength of the cluster formation, on the other. Empirical and computational probing is suggested that permits to test a fullerene-doped matrix for its applicability towards nonlinear optical applications.

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