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A many-particle quantum-kinetic formalism for describing properties of light emitters in frozen dielectrics

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Abstract. A many particle quantum-kinetic formalism is suggested to derive the Maxwell-Bloch-type equations which describe the interaction of quantum emitters with light in a frozen dielectric. It is shown that the quantum-kinetic formalism can meet the concept of local variations of dielectric properties and their influence on the emitter. The definitions of the local response and the effective refractive index in macroscopically homogeneous media are discussed.

We suggest a rigorous methodological approach that is essential in the theory of light emission by either a single quantum system or a light emitting ensemble when dynamics of the excited states and radiative properties are essentially dependent on its macroscopic dielectric host and the nearby surroundings. Most experimental data suggest that the properties of light from a single emitter are determined by the environment at the micro-, meso-, and macroscales. The corresponding analytical theories, however, remain either model-dependent or limited to describing specific situations and effects. Many studies found in the literature have been aimed at determining the change in the rate of the radiative decay of an excited single particle in a model environment as compared to its fluorescence in a material vacuum. These approaches are conventionally divided into "microscopic" and "macroscopic" models [1]. The common points in the models are the attempts to evaluate modifications of intrinsic properties of the emitting particle, the structure of the local driving field and the change in the local density of photonic states.

This research is focused on the properties of quantum light emitters embedded in a host material with likely local inhomogeneities in the presence of an external cw-laser beam. The mathematical tools used in laser physics, nonlinear and quantum optics, and other research related to interaction of radiation with the matter is largely based on an analysis of Maxwell-Bloch (MB) equations. To date, there has been a limited number of papers suggesting MB equations derived in a consistent manner and which include the effective values of all parameters: the Rabi frequency, transition frequency shifts and the rates of relaxation/excitation mechanisms (including spontaneous emission and dipole-dipole interactions) [1, 2]. In this work, we have made a joint between a sophisticated system of

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MB-type equations and observations of the excitations lifetime dispersion of single molecules (SM) in frozen matrices [3-5]. The lifetimes of the SM excited state can be measured using the data from SM zero-phonon line (ZPL) spectroscopy [6], at the conditions when the contribution from electron-phonon and electron-tunneling couplings are negligible. These conditions must be justified by providing extra accuracy in measuring ZPL power saturation and temperature dependences for each SM [6-9]. The excitation lifetime is a parameter in MB equations and must agree with the observations.

The MB-type system was derived for emitters in an ensemble of motionless particles embedded in a dielectric medium, which is transparent for the incident light. It includes the effective rates of individual and collective radiative damping, the Rabi frequencies, and frequency shifts of the transitions caused by the presence of the dielectric host and other quantum objects. These MB parameters were found to be functions of the real and imaginary parts of the host's permittivity. The excitation lifetimes of the emitters were shown to be functions of the effective refractive index and the extinction coefficient of the host around the emitter and agree with the experimental data.

Additionally, this work suggests a discussion of the results of the nanoscale mapping of the effective refractive index values in frozen dye-doped solutions performed by single-molecule spectromicroscopy [10,11]. It provides an estimation of the minimum volume of the host material which one may attribute to the effective refractive index.

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