Local and macroscopic characterization with single molecules and single quantum emitters

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Abstract. We introduce an experimental approach for mapping effective local values of dielectric characteristics of solid films and the analysis of the related local-field effects. The characterisation technique is based on the imaging and spectroscopy of single chromophore molecules at cryogenic temperatures. The progress in the theory of fluorescence enhancement due to the local field effects is reported.

We report the progress in development of the experimental approach for mapping effective values of local dielectric characteristics of frozen solid films and the analysis of the effects either enhanced or suppressed by the local fields. This probing technique is based on imaging and spectroscopy of single chromophore molecules at cryogenic temperatures. Since the fluorescence lifetime of a single dye molecule imbedded in a transparent matrix depends on the properties of the encapsulating medium at different spatial scales it has long been a challenge to detect inhomogeneities in the host material by simply measuring the lifetime of individual molecules distributed over the sample [1]. The factors that change the excitation lifetime in the matrix are known to be related to existence of the local density of the photon states and the local response of the medium to incident light. Both factors may be attributed to local effective values of the dielectric function describing the continuous surroundings of each chromophore light emitter. Thus, measuring the radiative lifetime distribution may help revealing the distribution of effective permittivities or refractive indices as well as the pattern of the local fields. In our experiment the spatial mapping of the local values is accomplished by localizing the corresponding chromophores with nanometer accuracy. We demonstrate this approach for a polycrystalline film doped with terrylene molecules [2]. We also report a significant progress in the theory of light emission enhancement due to the local field effects [3].

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