Measurement of Local Field Effects of the Host on the Lifetimes of Embedded Emitters

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We report experimental results on the variation of the radiative lifetime of Eu^{3+} ion embedded in a dielectric with the refractive index n. We dope 1 mol % of Eu^{3+} into the binary glass system xPbO-$(1-x)B_2O_3$. By varying x we have achieved a fairly large variation of the refractive index from 1.7 to 2.2. This enables us to study the local field effects for the first time for ions doped in a solid glassy material. Our measurements are in agreement with the so-called real cavity model. The present measurements are free from the complications arising from reorganizational effects in solvents.

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It was recognized very early by Lorentz that in a dense medium the field acting on an atom is neither the external field nor the Maxwell field, but an effective field that depends on the presence of all the other atoms in the medium [1]. Lorentz presented a very simple argument to show how such fields can be calculated. A microscopic approach to local fields was developed by Ewald and Oseen [1]. The local field effects have been very extensively studied in linear optics and in nonlinear optics and have been reviewed [2–5]. The effect of local fields on the propagation of light in multilevel resonant media has also been examined [6–12]. In a resonant dense medium one discovers very interesting frequency shifts and excitation dependent local fields [6,8,11]. The effects of local fields on electromagnetically induced transparency and coherent population trapping have been examined [8,10,12]. While a large body of theoretical literature exists, the experiments on the local field effects have been far too few [7,11,13].

We note that most studies deal with the propagation of light in a dense medium which could be either linear or nonlinear. A question of considerable importance in recent times concerns the modification of the decay rate of an atom (or ion) in a host material (such as a dielectric), particularly when the host is itself a dense medium. This is due to the fact that the total field at the site of the atom (ion) is altered by the presence of the dielectric. Since, according to the Fermi golden rule, the spontaneous emission probability is proportional to the square of the magnitude of the field at the location, the local field produced by the dielectric can change the lifetimes of the radiating ion relative to its value in free space or that in an optically thin medium. This problem has been a subject of rather extensive study in recent years. In these works the calculation of the local field is based on the assumption that the radiating dipole (ion) is placed in a spherical cavity, whose dimensions are large with respect to that of the radiating ion, but small compared to the wavelengths involved. The radiative lifetime of the emitter embedded in the dielectric can then be expressed as $\tau(n) = \tau(0)l(n)/n$. Here $l(n)$ is the local field correction factor (LFCF), $\tau(0)$ is the free space radiative lifetime, and n is the refractive index at the wavelength of emission. In the absence of the local fields, the radiative lifetime follows a simple inverse relationship to the refractive index [14], i.e., $\tau(n) = \tau(0)/n$. One has to then determine the form of $l(n)$, which requires specification of the nature of the cavity. Two distinct models for the nature of cavities—real and virtual—have been proposed in the literature. The LFCFs, l, for the real and virtual cavities are $(2n^2 + 1)^2/9n^4$ and $9/(n^2 + 2)^2$, respectively [2,15,16].

In the real cavity model it is assumed that the atom is at the center of a cavity and that the cavity itself has no other material. The virtual cavity model is based on the work of Lorentz [2]. It is *a priori* not clear which model is more relevant for a given experimental situation. The existing experimental works were carried out either in liquids [17,18] or a gaseous [19] medium. However, the range of refractive indices obtainable in gases and liquids is not only small, but also the method used to obtain the refractive index variation is prone to complications. For example it is known that in a solution the solvent molecules tend to reorganize in the vicinity of the metal ion, thereby leading to a local dielectric constant, which could be different from the bulk value [20]. The choice of the host material and the emitters is very critical in understanding the local field related issues. Ideally a far better host material will be a solid matrix.

In this Letter, we report experimental results on the decay of europium ions in specially prepared glasses. We explain later the advantages of our choice of dopants and the host material. We are able to cover a wide range of the refractive index of the host material. Moreover our results are free from the complications of the gaseous or liquid host materials. Our data support the real cavity model. A

