Experimental studies of spontaneous emission from dopants in an absorbing dielectric

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We report the first measurements, to our knowledge, of the modification of spontaneous emission rates of Eu$^{3+}$ ions in the visible region owing to an absorbing medium. Precise levels of the absorption coefficient are introduced by codoping with different amounts of Nd$^{3+}$. We use a binary glass system PbO–B$_2$O$_3$ as the host, the compositional variation of which leads to a change in the real part of the refractive index. Measured lifetimes are found to follow the real cavity model, and the data are analyzed by the model proposed by Scheel et al. [Phys. Rev. A 60, 4094 (1999)]. We give estimates of the parameter that is related to the radius of the cavity around Eu$^{3+}$. © 2005 Optical Society of America

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The radiative lifetime of an atom in a dielectric is not only determined by the electronic wave functions of the atomic states involved but also by the photon density of states and the electromagnetic field strength of the photon modes at the position of the atom. The spontaneous decay rate of an atom placed in a dielectric can be modified when compared with its free-space rate, given by

$$\Gamma_0 = \frac{\mu^2 \omega^3}{3\pi\varepsilon_0 c^3},$$

where $\mu$ is the dipole moment and $\omega$ is the transition frequency. Modification of the spontaneous emission rate has been studied for various systems such as atoms embedded in a dielectric, free atoms in resonant cavities, organic molecules, Eu$^{3+}$ complexes in solution, III–V semiconductor quantum wells, and Er$^{3+}$-doped SiO$_2$. Rikken and Kessener demonstrated the modification of lifetimes of Eu$^{3+}$ complexes dissolved in different solvents and Shuurmans et al. demonstrated the same in Eu$^{3+}$ in supercritical CO$_2$ gas. The lifetimes in the presence of a bulk and transparent dielectric can be written as

$$\tau(n) = \frac{\tau_0}{n},$$

Here $\tau(n)$ is the lifetime in the medium of (real) refractive index $n$; $\tau_0$ is the free-space lifetime; and $l(n)$ is the local field correction factor, which in addition is also found to give rise to frequency shifts in a two-level system. While calculating the local field, the atom is assumed to be at the center of a spherical cavity. The dimensions of such a cavity are assumed to be large compared with the dimensions of the atom and small compared with the wavelengths involved. Two distinct models, real and virtual for the nature of the cavities, have been proposed. The $l(n)$ for real and virtual cavities are $(2n^2+1)^2/2n^4$ and $9/(n^2+2)^2$, respectively. In the real-cavity model it is assumed that the atom is at the center of the cavity and the cavity itself has no other material. The virtual-cavity model is based on the work of Lorentz and assumes a uniform distribution of material within and outside the cavity. In a recent experiment we measured the lifetimes of Eu$^{3+}$ in bulk PbO–B$_2$O$_3$ glass. Our measurements were found to agree well with the real-cavity model.

In many real situations the radiating ion would be surrounded by an absorbing medium. Such a situation may arise from self-absorption, impurities, codopants, etc. In such situations the refractive index is no longer real and one has to take into account the complex nature of the refractive index of the medium. An exact theoretical treatment of the spontaneous decay of an excited two-level atom in the presence of a dispersive and absorbing dielectric medium was presented by Scheel et al. Experimentally this situation can be created by suitably doping the radiating ion in an absorbing medium. Because of the well-known energy level structure of rare-earth elements, one can choose two rare-earth elements in such a way that the absorption of one of the elements overlaps the emission of the other. The energy transfer from Eu$^{3+}$ to Nd$^{3+}$ has been studied in various hosts, including glasses. PbO–B$_2$O$_3$ glass as a host is attractive not only because it is easy to fabricate but also for its application in wavelength-division multiplexing.

No experimental studies are available in the literature that take into account the effects of both the real and the imaginary components of the refractive index of the dielectric on the rates of spontaneous emission of an atom embedded in a medium. Here we present the first estimate, to our knowledge, of the spontaneous emission rates of Eu$^{3+}$ ion embedded in a dielectric of PbO–B$_2$O$_3$ with different concentrations of Nd$^{3+}$ as a codopant. Because the absorption of Nd$^{3+}$ overlaps with the emission spectrum of Eu$^{3+}$ and both Eu$^{3+}$ and Nd$^{3+}$ can be introduced into the binary complex, this system seems to be an excellent probe for characterizing the spontaneous emission rates for dopants in an absorbing and dispersive dielectric medium.

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