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Local field effects on the radiative lifetime of emitters in surrounding media: Virtual- or real-cavity model?

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Abstract

By close analysis of the available experimental results on the spontaneous emission lifetime of various emitters in different media and examining the assumptions underlying the two titled models, we reach a consistent interpretation of the experimental results and give a practical criterion on which model is applicable for a given situation.

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It has long been known that the spontaneous emission lifetime of emitters can be modified by changing the surrounding dielectric media [1,2]. The theory on this subject continues to attract considerable attentions due to its fundamental importance and its relevance to various applications in low-dimensional optical materials and photonic crystals [3–5]. Various macroscopic (see Ref. [2] for a recent review) and microscopic [5–8] theoretical models have been developed to model the dependence of the spontaneous emission rates (or lifetimes) on refractive index. Among those models are the real-cavity model (also referred to as empty-cavity model [2]), where emitters (usually ions) are assumed to create tiny cavities when replacing host ions, and the virtual-cavity model, which is based on the Lorentz local field [7]. Different models predict substantially different dependences of radiative lifetime on the surrounding media. There are also some measurements intended to discriminate these models, with most experimental results tend to agree with the real-cavity model. Especially, recent measurements on the

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radiative lifetime of Eu^{3+} ion embedded in glass of varying refractive index [4] also tend to agree with the real-cavity model, in contrary to the general belief that the virtual-cavity model should be more relevant. Duan and Reid [9] pointed out that $4f \to 4f$ electric dipole radiative relaxation, which is due to mixing in $4f^N$ states with states of opposite parity, depends strongly on the environment and does not serve as a good examination of the two models. To overcome the problem in $4f \to 4f$ transitions, Duan and Reid [9] analyzed the lifetimes of $5d \to 4f$ transitions of Ce^{3+} ions in hosts of different refractive indices and the results maintained the text-book virtual-cavity model. However, there was no intention to answer the questions why most other experimental results agree with the real-cavity model, and which model should be used for a given situation. Early theoretical work by de Vries and Lagendijk [6] gave a criterion on which model should be applied by distinguishing the case of emitters as "interstitial" impurities, where the Lorentz model is applicable, from the case of emitters as "substitutional" impurities, where the real-cavity model is relevant.

In this Letter, we will examine those reported experimental results on various emitters in different surrounding media that appear to support different models, point out the problems with some previous interpretations, and reinterpret those experimental results with more reasonable models that consistent with the local structure of emitters in the media. Finally we answer the titled question by our consistent explanation of the experimental results and propose a practical criterion on finding out the proper model for a given situation.

Measurements on the radiative lifetime of Eu^{3+}-hfa-topo complex emitter in a series of apolar hydrocarbons was reported by Rikken et al. in 1995 [10]. Experimental results show that the emissions are from $^5D_0$ levels and dominated by electric dipole transitions to $^7F_2$ levels, and the radiative quantum yield is close to unity. Fig. 1 is the plot of the measured lifetimes together with the best fitting using the real-cavity model:

$$\tau_{\mathrm{real}} = \tau_0 \frac{1}{n\left(\frac{3n^2}{2n^2+1}\right)^2}, \tag{1}$$

where τ_0 is the lifetime of the emitter in vacuum, the factor n is due to the reduction of speed of light in the media and $(\frac{3n^2}{2n^2+1})^2$ is square of the ratio of the electric field in the cavity (local field) to the macroscopic field in the media. τ_0 is taken to be an adjustable parameter independent of the refractive index in the fitting.

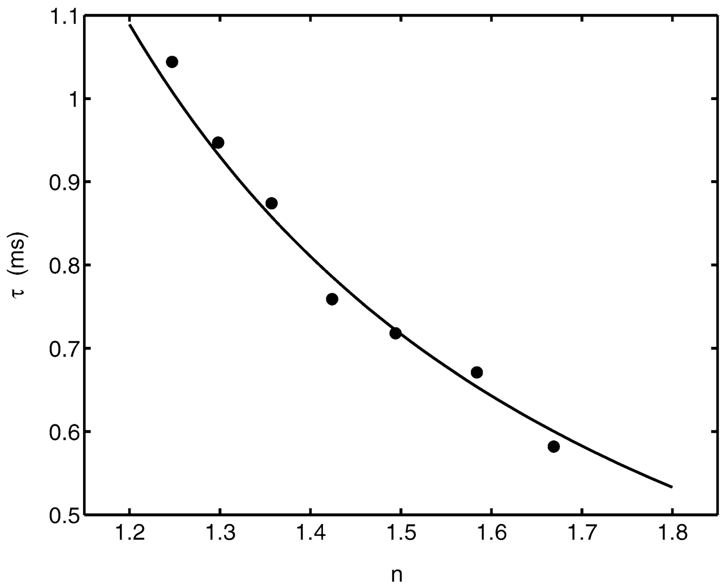

Fig. 1. Radiative lifetime of Eu^{3+}-hfa-topo as a function of the solvent refractive index. The dots are measured lifetimes and the curve is fit to Eq. (1).