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# Plasmon-enhanced fluorescence in nanomolar dye solution using combination of core-shell nanostructures of various shell thicknesses

R. Paul<sup>1,2</sup>, A. Arya, R. Laha<sup>1</sup>, V. R. Dantham<sup>\*1</sup>, S. Hussain<sup>2</sup>

<sup>1</sup>Department of Physics, Indian Institute of Technology Patna, Bihar, India – 801103

<sup>2</sup>Department of Chemistry, Indian Institute of Technology Patna, Bihar, India – 801103

\*Corresponding author: dantham@iitp.ac.in

## ABSTRACT

Enhancement in fluorescence of samples in solution phase is very crucial for many practical applications. Herein, we report the fluorescence enhancement of a nanomolar solution using Ag core-silica shell nanostructures synthesized independently. Coumarin 6H dye solution has been taken as a case study. Three different Ag core – silica shell nanostructure solutions were prepared by varying the amount of reagent in Stöber method. The core-shell nanostructures were characterized by atomic force microscopy, dynamic light scattering, and UV-Visible spectrophotometry. The maximum values of fluorescence enhancement ( $\eta_{max}$ ) were found to be 2.5, 4.2, and 1.5 when respective solutions with average nanoshell thicknesses of 2.4, 16.7 and 28.3 nm were used individually. However, the value of  $\eta_{max}$  increased by an order ( $\eta_{max} \sim 13$ ) when the mixture of the nanoshell solutions was used. The reason behind the improvement in  $\eta$  was ascribed to the optimization in the values of the distance ( $r$ ) between nanoshell and dye molecules as well as the separation ( $d$ ) between the nanoshells. This report, as we believe, will enable the scientific community in obtaining better enhancement in the fluorescence signal of labeled proteins and fluorescent molecules in aqueous media with ease.

Keywords: Surface plasmons; Plasmonics; Fluorescence; Nanostructure fabrication; Subwavelength structures, nanostructures

## 1. Introduction

Sensing and imaging of biomolecules with different dye labels have created a lot of interest recently in biochemistry and molecular biology [1-4]. In both cases, the intensity of fluorescence from the dye labels determines the sensitivity and imaging brightness. Due to this, researchers have been trying to improve fluorescence signal using the technique called as surface enhanced fluorescence (SEF) which is also known as metal enhanced fluorescence in some section of literature [5, 6]. The signal enhancement is obtained in SEF by placing the labels close to the nanoplasmonic structures of various sizes and shapes. The SEF phenomenon is a two-step process namely (I) enhancement of local field due to the excitation of localized surface plasmons of metal nanostructures and (II) coupling between local field and molecular dipole of the fluorescent label [7]. According to the radiating plasmon model, nanoplasmonic structures can both enhance and quench the fluorescence at the location of the fluorophore, depending upon the separation between the nanoplasmonic structure and the fluorophore [8]. If the separation is too small, then the fluorescent molecules in the excited state would generate the non-radiative energy which is transferred to the metal nanoparticles. This results in weakening or even quenching the fluorescence. On the other hand, fluorescence molecules would not be affected by local field of nanoplasmonic structure for larger separation. Hence, the significant fluorescence enhancement could be achieved only when the separation is an optimum one, which again depends on the size of the metal nanoparticle. This optimum separation is typically about 5–20 nm.

In order to keep separation between fluorescent molecules and nanoplasmonic structures, DNA [9, 10] and different polymer materials have been used [11-13] as spacers. However, the disadvantage of using DNA and polymers as spacers is that due to their flexibility, these structures don't allow a complete control of the metal-dye distance and can even fail to prevent dye-metal contact, leading to emission quenching [14]. To overcome this problem, silica material is used as a spacer by coating plasmonic structures with silica material. The silica material is preferred because it has anomalously high stability especially in aqueous media, chemical inertness, controlled porosity, process ability, and optical transparency [15]. Additionally, this coating should endow the metal cores with several beneficial properties, such as the possibility of subsequent surface modification/functionalization and biocompatibility [15].

One of the initial demonstrations of the fluorescence enhancement using metal core – dielectric nanoshells was by Hooisweng *et al.* [16]. Later on, several researchers have demonstrated the enhancement of fluorescence of different fluorophores [17-27]. Zhang *et al.* have demonstrated successfully the upconversion fluorescence using core-shell nanoparticles [28]. Liaw *et al.* have developed analytical theory to estimate the average fluorescence enhancement of molecules doped core-shell nanoparticles using Mie theory and dyadic Green's function [29]. These nanoparticles are also found better in pH sensing and individual bacterial detection [30, 31]. Ribeiro *et al.* have demonstrated a new methodology for the fluorescence enhancement free from inner

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