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Numerical investigation of surface plasmon resonance effects on photocatalytic activities using silver nanobeads photodeposited onto a titanium dioxide layer

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ABSTRACT

We numerically investigated the surface plasmon resonance effects on photocatalytic activities using silver nanobeads photodeposited onto a titanium dioxide (TiO₂) layer by three-dimensional finite element method. Results show that the proposed case C structure exhibits significantly high photocatalytic activity in a wide range of incident angle of light and broad wavelength range in near-UV, visible and near-infrared. The enhanced electric field region, intensity and peak resonance wavelength could be increased by enhancing the inner medium dielectric constant (ϵ) in silver nanobeads with ϵ in the range 1.7689–11.56 and the incident angle of light θ in the range 30°–75°.

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1. Introduction

Considerable research on photocatalysis during the recent years continues to provide fundamental insights and practical applications [1–7]. Various principles and measurements have been obtained with photocatalysis, where the strong photooxidative activity of titanium dioxide (TiO₂) has been applied to environmental cleanup [3,4]. It will be a great benefit to apply photocatalysis to treat low-concentration pollutants in air and water, and to decompose pollutants adsorbed on TiO₂ surfaces [8–11]. This resulted in the concept of “light cleaning,” i.e., deodorizing, disinfection, and decontamination of air, water, and surface with TiO₂ layers and light [5,12].

It is well known that TiO₂ displays photocatalytic behavior under near-ultraviolet (UV) illumination. The photocatalytic activity of TiO₂ has been markedly improved through efforts of many research groups [12–20]. TiO₂ is recognized as a fascinating material that shows photoelectrochemical solar-energy conversion. TiO₂ in anatase phase has been widely used as a conventional photocatalyst [12]. Applications of TiO₂ extend into various areas of photocatalysis [12–14], including self-cleaning surfaces and

photoinduced superhydrophilicity [12,15]. In another scientific field, the excitation of localized plasmon polaritons on the surface of silver (Ag) nanoparticles causes an enormous increase of the near-field amplitude at well-defined wavelengths in the near UV [12]. The exact peak resonance wavelength (λ_{res}) depends on the shape and the dielectric environment of the metal nanoparticles (MNPs). It is expected that the photocatalytic behavior of TiO₂ would be greatly boosted if it gets assisted by the enhanced near-field amplitudes of localized surface plasmon resonance (SPR). This phenomenon is termed as “plasmonic photocatalysis” [12]. The idea of plasmonic photocatalysis is as follows. TiO₂ in anatase phase is a semiconductor with a band gap of 3.26 eV, so near-UV irradiation can excite pairs of electrons and holes. Ag nanoparticles show a very intense localized SPR absorption band in the near-UV region. This is associated with a considerable enhancement of the electric near-field in the vicinity of the Ag nanoparticles. We therefore hypothesized that this enhanced near-field could boost the excitation of electron–hole pairs in TiO₂ and therefore increase the efficiency of photocatalysis.

Significant advances have been made in recent years to design metal core–semiconductor shell clusters [12,16–23]. Yet, the efforts to utilize these core–shell structures (e.g., SiO₂ coated on solid Ag) as photocatalysts in the light energy conversion systems (e.g., photoelectrochemical cells, hydrogen production) are limited. The optical properties of different types of plasmonic

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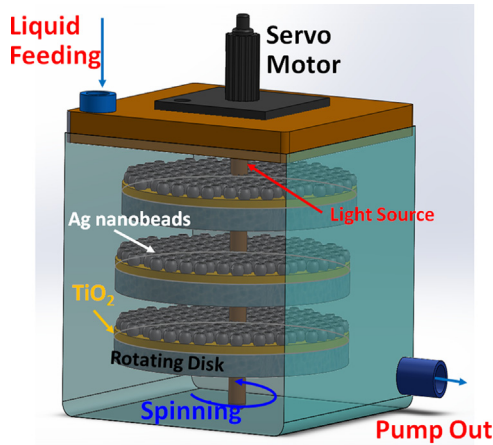


Fig. 1. Schematic diagram of Ag nanobeads photodeposited onto a TiO₂ layer.

photocatalysis by using MNP (e.g., Au, Ag) coated by SiO₂ have been discussed over the last few years [5,10,12–16]. However, the effects on metallic nanobead plasmonic photocatalysis have not been considered. As indicated in our previous works, the effects of SPR of Ag nanoshell are superior to that of solid Ag in nanoscale [24]. Nanoshells (or nanobeads) possess several attractive features which make them interesting as nanoscale optical components [25]. Additionally, it has been demonstrated that nanoshells and other nanoscale MNP structures greatly enhance local electromagnetic fields in certain regions near their surfaces at specific wavelengths of light, controlled by nanostructure geometry [26]. This subwavelength structure could provide a tool for manipulating light below the diffraction limit.

The optical properties of Ag nanoparticles are dominated by SPR effects, defined as the collective motion of the conduction electrons induced by light irradiation [27,28]. In order to obtain efficient photocatalysts that work under solar irradiation, in this work we propose a plasmonic photocatalyst reactant coated on a TiO₂ layer [5], i.e., Ag nanobeads were periodically photodeposited onto the surfaces of TiO₂ layer as shown in Fig. 1(a). All Ag nanobeads showed localized SPR in the range of near-UV, visible and near-infrared, indicating the presence of Ag nanobeads with different incident angles of light. We have compared the optical response in near-field zone with and without the Ag nanobead on the surface of TiO₂ layer. The influences of incident angle of light and filling medium in DHs on local field enhancement in the vicinity of TiO₂ surface are elaborated.

2. Simulation method and models

All of the simulated spectra, electric field intensities, energy flows and charge densities are obtained by solving three-dimensional (3-D) Maxwell equations with a finite element method (FEM) [29,30] in 3-D calculations. FEM was used to solve the Helmholtz equation in 3-D spatial domain for simulation models as shown in Fig. 2(a)–(c). For simulating the periodic array of MNPs in water we set periodic boundary condition (PBC) along the side walls to mimic the behavior of an infinite periodic array, thereby including some coupling effects between neighboring MNPs. Perfectly matched layer (PML) boundaries are set on the top and bottom surfaces to suppress any reflection from boundaries of computation window in order to save the computer resource and simulation time. By calculating the absorption, we can use the scattering parameters (*S*-parameter) and obtain the

absorption spectrum:

$$A \text{ (absorption)} = 1 - R \text{ (reflection)} - T \text{ (transmittance)} \quad (1)$$

The definitions of *S*-parameters, *S*₁₁ and *S*₂₁, are

$$S_{11} = \left[\frac{\text{power reflected from port 1}}{\text{power incident on port 1}} \right]^{1/2} \quad (2)$$

$$S_{21} = \left[\frac{\text{power reflected from port 2}}{\text{power incident on port 1}} \right]^{1/2} \quad (3)$$

The electromagnetic wave is incident on port 1 and port 2 in a receiver plane [30]. Reflectance and transmittance are calculated by $|S_{11}|^2$ and $|S_{21}|^2$, respectively, where port 1 and port 2 are indicated in the inset of Fig. 2(a)–(c).

The peak wavelength of the SPR is sensitive to both the MNP size and the medium surrounding the MNP. The 3-D FEM that we applied to MNPs was comprehensively described in Refs. [29–32]. Throughout this paper, we consider Ag as nanoparticles/nanobeads, illuminated by a transverse magnetic (TM) plane wave. The periods (Λ) along *x* and *y* axes are $\Lambda = 150$ nm. For the Ag permittivity $\epsilon(\lambda)$ as a function of wavelength (λ) we use the bulk data of Johnson and Christy [33]. Refractive indices of the dielectric materials were taken from literature [34].

For experimental realization, Ag nanobeads were fabricated by employing a multistep process similar to other recently developed fabrication procedures [35–37] to have a 35 nm radius air (or dielectric) core and a 15 nm thick shell. A solution of the photocatalytic TiO₂ film of thickness of 10–40 nm was spin-coated onto the SiO₂ layer [10,12]. The thick SiO₂ is opaque like a mirror. Thus an appropriate thickness of the TiO₂ layer in order to have an overall high absorption is estimated to be less than 100 nm. In other words, it is impossible to enhance photocatalytic activity with thick TiO₂ [12]. We choose the TiO₂ layer with a thickness of 40 nm in our work.

Fig. 1 shows a schematic diagram of Ag nanobeads photodeposited onto a TiO₂ layer [5]. The unit cell of simulation models is shown in Fig. 2(a)–(c), i.e., (a) a unit cell of TiO₂ photocatalyst system without MNPs (termed as case A hereafter), (b) a unit cell of periodic array of solid Ag nanoparticles with diameter of 50 nm photodeposited onto a TiO₂ layer (termed as case B hereafter), and (c) a unit cell of periodic array of core–shell Ag nanobeads with shell-thickness of 15 nm photodeposited onto a TiO₂ layer (termed as case C hereafter). From our point of view, the proposed plasmonic photocatalyst system as shown in Fig. 1 can effectively treat pollutants in air or water to yield an efficient photocatalyst with ability of working under solar irradiation. The layer structure from bottom to top is SiO₂ ($n = 1.5$, $h = 310$ nm, where n is refractive index and h is the thickness of layer)/TiO₂ ($n = 0.076 + i1.606$, $h = 40$ nm)/Ag nanoparticle (or nanobead) ($n = 1.620 + i0.200$, outer diameter of 20 nm for Ag nanoparticle)/water ($n = 1.33$)/air ($n = 1$, $h = 175$ nm). We have assumed water as the surrounding medium in the entire outer space. An individual Ag nanobead as shown in case C [see Fig. 2(c)] is chosen with the outer diameter of 50 nm and inner diameter of 35 nm (i.e., shell thickness 15 nm). We assume that the Ag nanoparticles (see Fig. 2(b)) and the Ag nanobeads (see Fig. 2(c)) are of the same size and periodically distributed (period Λ) on the surface of TiO₂ layer as shown in Fig. 2(d). In order to obtain efficient photocatalysts with incident light in the range of 300–1200 nm (i.e., ranging in near-UV, visible and near-infrared) interacting with the proposed system, the simulation models shown in Figs. 2(a)–(c) are illuminated by oblique incidence light wave with TM polarization. The parameter θ is defined as the angle of incident light to explore the variation of field enhancement on the surface of TiO₂ layer.

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