## Accepted Manuscript

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PII:	S1386-1425(18)30490-6
DOI:	doi:10.1016/j.saa.2018.05.089
Reference:	SAA 16123
To appear in:	Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy
Received date:	4 February 2018
Revised date:	22 May 2018
Accepted date:	27 May 2018

Please cite this article as: Fengjiao Zhao, Guanghua Ren, Canhua Zhou, Yang Yang, Study of one-step photosynthesis of Ag nanoparticles. Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy(2017), doi:10.1016/j.saa.2018.05.089

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### **Study of One-Step Photosynthesis of Ag Nanoparticles**

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#### Abstract

During photosynthesis of metal nanoparticles (NPs) in aromatic ketone systems, the aromatic ketone triplets are explored as hydrogen atom/electron abstractors which yield highly reactive radicals. However, their electron donation ability is sparsely recognized. Here we report the investigations of benzophenone (BP)-initiated one-step photosynthesis of Ag NPs in H donor free system using spectroscopic and theoretical methods. Experimentally, the direct observations of BP cation radical (BP<sup>++</sup>) and Ag NPs *via* transit and steady-state absorption spectroscopy demonstrate the electron donation ability of BP triplets which directly reduce Ag<sup>+</sup> to Ag<sup>0</sup>. In the following reactions, both theoretical calculation and experimental results reveal that the generated BP<sup>++</sup> recovers itself to BP by grabbing an electron from NO<sub>3</sub><sup>-</sup> and the generated nitrate radicals (NO<sub>3</sub><sup>-</sup>) then decay to nitrogen oxides. Notably, BP plays a role as photocatalyst in our system. This study details the mechanism of one-step photosynthesis of Ag NPs which provides a new insight of aromatic ketones in photochemistry and may offer a facile approach to photosynthesis of metal NPs.

**Keywords:** photosynthesis, transient absorption spectroscopy, dynamics, metallic nanoparticles, benzophenone cation radical, electron transfer, electron donation

#### 1. Introduction

Noble metal NPs is in great demand [1-2] because of their excellent properties and wide use in energy production [3-5], biological molecules detection [6-8] and medical treatment [9-11]. Due to surface plasma resonance effect, spectroscopic method has been used to detect noble metal NPs by measuring surface plasmon band (SPB) absorbance. As a popular class of photosensitizer for metal NPs photosynthesis, aromatic ketones have attracted extensive attention for strong absorption in ultraviolet [12-17] and high efficiency of intersystem crossing (ISC) [18-20]. Moreover, the characteristic of efficient photon-induced electron transfer (ET) [21-24] makes aromatic ketones a preferred photoinitiator in photochemistry field. From previous studies, with the presence of hydrogen atom/electron donors (eg: ethanol, trimethylamine, polyvinyl pyrrolidone, etc), aromatic ketone triplets exhibit strong hydrogen atom/electron abstraction ability [1-3,25-29]. The generated highly reactive radicals after hydrogen atom/electron transfer are favorable specials in photosynthesis of metal NPs. To explore and improve radical's performance and generation quantum yield, most works focused on their electron/hydrogen atom abstraction property. As a result, electron donating ability of aromatic ketone triplets is barely recognized and reported.

Benzophenone (BP) which is the typical and peculiar aromatic ketone photosensitizer, has simple structure and extraordinary high efficiency of ISC ( $\Phi_{ISC}$ ~1) [18]. This character makes BP especially suited for triplet state study using ultrafast transient absorption (TA) spectroscopy. In

2005, Farid and co-workers described BP triplets as electron donor in study of chain-amplified photoreactions and assigned TA peak at 390nm to BP<sup>++</sup> for the first time. [30] Later in 2013, Scaiano's group reported the potential role of BP triplets as reducer in the system containing H<sub>2</sub>O. [31] This makes electron donation property of BP triplets in H donor-free systems for metal NPs synthesis remains unclear.

In this work, combining experimental and theoretical methods, we systematically studied the mechanism of BP-initiated one-step photosynthesis of Ag NPs in simple system of BP with AgNO<sub>3</sub>. AgNO<sub>3</sub> was chosen for its suitable reduction potential which is  $E_{Ag+}=0.799V$  at 20°C. [32] To eliminate possible effect of H<sub>2</sub>O as an H donor, all samples are prepared in super-dry acetonitrile (ACN). After excitation (@355 nm), BP goes through highly efficient ISC process and yields BP triplet (BP T<sub>1</sub>). Electron donating process of BP T<sub>1</sub> was quantified using nanosecond TA spectroscopy in which the formation and decay processes of BP<sup>++</sup> were directly detected. Silver NPs formation was explored *via* UV/Vis

**Scheme 1.** Benzophenone structure and Ag NPs photosynthesis processes in BP-AgNO<sub>3</sub> system.



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