

## Theoretical analysis of the role of triplet-triplet annihilation in triplet-state-based optical manipulation

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Since Ashkin's pioneering work on the trapping of microparticles using a strongly focused beam, optical manipulation has been extensively studied. However, manipulating nanoscale materials that are small and interact weakly with light poses challenges that require further enhancement of optical forces. One promising approach to address this challenge is the optical resonance effect (ORE), which increases optical forces by illuminating the material with light resonant with the excitation energy of a material's excited state [1]. While the application of ORE has traditionally been limited to singlet states, recent experimental successes in utilizing ORE in triplet states have been reported, resulting in a significant increase in the efficiency of optical manipulation [2]. Specifically, the study focused on Phenalenone molecules embedded to polystyrene spheres, with their energy-level structure is illustrated in Figure 1. States  $|1\rangle$  and  $|3\rangle$  correspond to the ground and excited states of the singlet states, respectively, while states  $|2\rangle$  and  $|4\rangle$  correspond to the ground and excited states of the triplet states. A key feature of this system is that triplet-state electrons are efficiently quenched by oxygen in the medium, rapidly relaxing to the ground state. Using a chemical stimulus to reduce the oxygen concentration and extend the lifetime of the triplet state  $|2\rangle$ , we demonstrated for the first time an enhancement of ORE, further improving the efficiency of optical manipulation.

In a previous study, we reported qualitative agreement between experimental results and theoretical predictions for this mechanism [3]. However, that study provided only a phenomenological explanation for the observed decrease in optical force under high-intensity excitation light, which was attributed to the saturation of triplet-state electron populations [4]. In this presentation, we hypothesize that this decrease is caused by triplet-triplet annihilation (TTA) and propose a new theory for optical manipulation incorporating TTA effects. The new theory explains the reduction in triplet-state electron populations and the corresponding decrease in optical force, which could not be captured by conventional theories. Building on these insights, we will also discuss changes observed when switching the light source from a CW laser to a pulsed laser, proposing a more efficient optical manipulation method. This study provides crucial insights for further enhancing the efficiency of ORE-based optical manipulation using triplet states.

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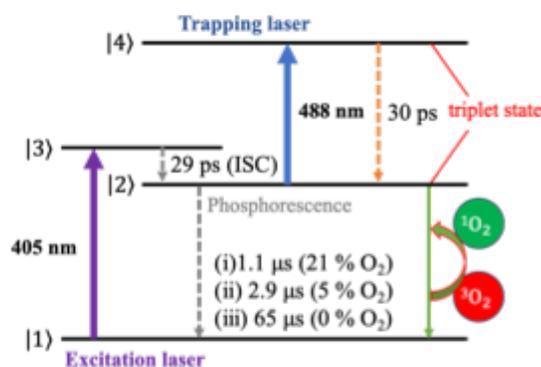


Fig. 1 Energy level structure of Phenalenone