Photo-Induced Triplet Depletion Allowing Higher-Resolution Afterglow

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Stimulated emission depletion microscopy uses stimulated emission from the singlet excited state of a chromophore, which allows super-resolution fluorescence images.[1] However, fluorescence imaging is often interfered by emission from surrounding environment in the presence of fluorescent impurities. As an imaging technique independent of fluorescent impurity, afterglow imaging using persistent room-temperature phosphorescence (RTP) has been reported.[2] However, no possible stimulated emission from the lowest triplet excited state (T_1) precludes us to make higher resolution-afterglow emission images.

Here, we demonstrate photoinduced triplet depletion and improved resolution of afterglow emission using the depletion. After ceasing excitation, persistent RTP from an amorphous *β*-estradiol film doped with 1 wt% chromophore **1** (Figure 1a) was depleted by the strong photoirradiation with a longer wavelength than an absorption wavelength of **1** (depletion beam) (Figure 1b). Triplet depletion possibility depending on irradiance of different depletion beam-colors indicates that a photoionization from the lowest unoccupied molecular orbital (LUMO) in T_1 over vacuum level of **1** triggers the triplet depletion ((i) in Figure 1c). Analyses of charge generation and upconversion emission behavior by irradiation of the depletion beam indicate that triplet depletion mechanism includes a process in which electrons eventually return to the highest occupied molecular orbital (HOMO) of **1** ((ii) in Figure 1c). By simultaneously focusing excitation and specially modulated depletion beam onto the film, a higher resolution afterglow emission was observed due to the triplet depletion (right in Figure 2).

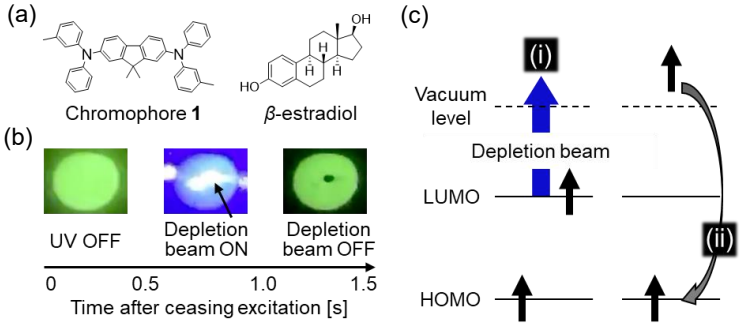

Figure 1. (a) Chemical structures. (b) Photographs showing RTP depletion. (c) Energy diagram illustrates RTP depletion mechanism.

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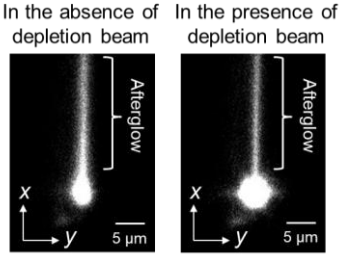

Figure 2. High-resolution afterglow imaging. Left and right show in the absence and presence of donut-shaped depletion beam, respectively.