

Article

# Optical dynamic nuclear polarization of $^{13}\text{C}$ spins in diamond at a low field with multi-tone microwave irradiation - Supplementary Material

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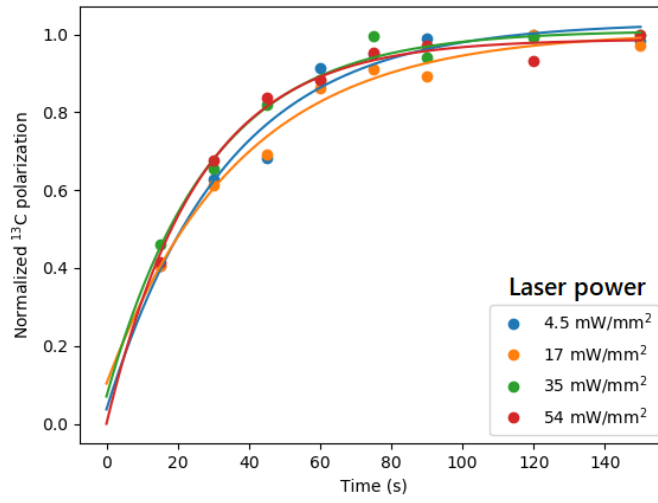
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### 1. $T_1$ comparison at different laser power

Spin-lattice relaxation time ( $T_1$ ) may significantly change as the diamond temperature increases by the laser-induced heating. But, it is hard to access directly to the temperature dependence of  $T_1$ . Instead, one can expect that the polarization time should reflect the spin-lattice relaxation time. Thus, we have measured polarization build-up curves at different laser power densities used for the optical DNP (Fig. 1). As shown in Fig. 4 (b) in the main text, the diamond temperature increases linearly with the laser power density. We have found that the polarization times are nearly identical up to the power density of 54 mW/mm<sup>2</sup>. This result indicates that the nuclear spin-lattice relaxation rate has almost no influence on the decrease of <sup>13</sup>C polarization shown in Fig. 4 (a) in the main text, and it supports our claim of the existence of additional depolarization process that is thermally activated by the laser-induced heating.



**Figure S1.** Normalized <sup>13</sup>C polarization vs build-up time at different laser power density.