Supporting information

SI-1. Absorption spectra of Azo PBD and AzoTATA

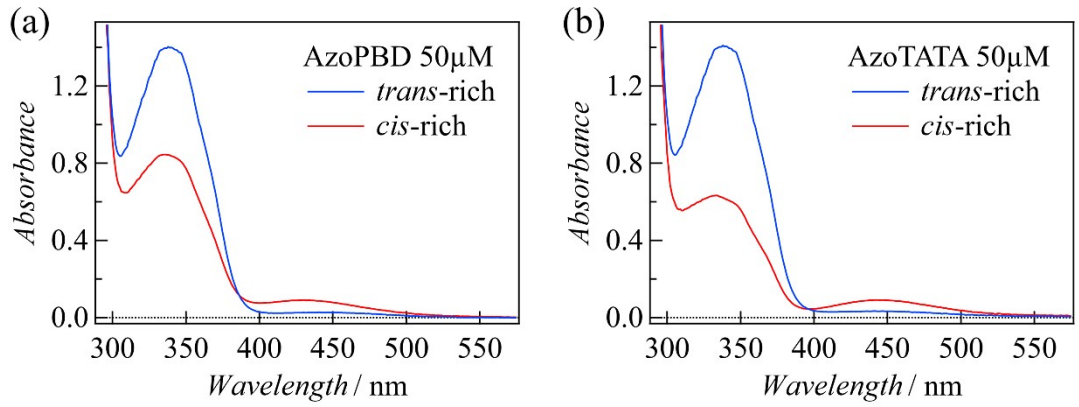

Figure S1. The absorption spectra of (a) AzoPBD and (b) AzoTATA upon blue (*trans*-rich) and UV (*cis*-rich) light illumination.

SI-2. TG signal of AzoPBD in the *trans* state

The steady state of azobenzene upon continuous UV light illumination should be a mixture of the *cis*- and *trans*-forms, because both forms possess UV-light absorption bands. Furthermore, since the *trans*-form has weak absorption in the blue light region, the observed TG signal upon blue light excitation should contain two contributions, *cis* → *trans* reaction and *trans* → *cis* reaction, simultaneously. To estimate the TG signal due to the *trans* → *cis* reaction, we measured the TG signal for the *trans*-rich solution by pre-illuminating with a blue LED (450 nm). The *trans*-rich solution was excited by a blue pulse for the TG measurement (Fig.S2). It was found that the intensity of the diffusion signal of the *trans*-rich solution was almost negligible (less than 0.3%) compared with the signal of the *cis*-rich solution. This result indicates that the observed TG signal described in the main text comes from the *cis* → *trans* reaction.

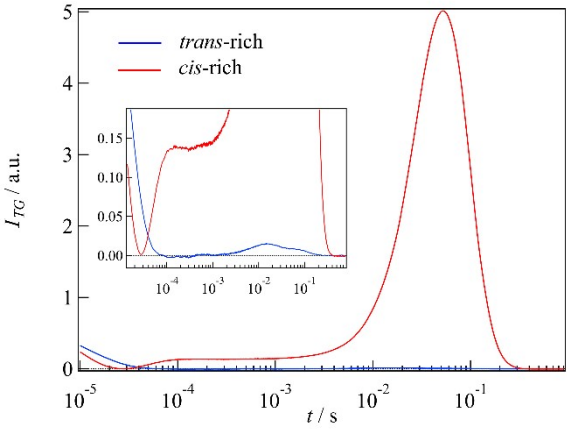

Figure S2. Comparison of TG signals between (red) *cis*-rich and (blue) *trans*-rich states of AzoPBD with T7 RNAP at q^2 of 3.2×10^{11} m^{-2}.

SI-3. Fitting function of TG signal of AzoPBD-T7 RNAP solution

The time development of the TG signal was analyzed based on the reaction model of Scheme 1 in the main text. By solving a diffusion-reaction equation, the time profile of $\delta n_{spe}(t)$ for Scheme 1 is obtained as follows,

$$\begin{aligned}\delta n_{spe}(t) =& -\delta n_R exp\left(-D_R q^2 t\right) + \left[\delta n_{I1} - \delta n_{I2}\{k_1/(k_1-k_2)\} + \delta n_{P_1}\{k_1k_2/(k_1-k_2)\}\left\{1/\left(D_{I1}-D_{P_1}\right)q^2 + \right.\right. \\ & exp\{\left(-D_{I1}q^2 + k_1\right)t\} \\ & + \left[\delta n_{I2}\{k_1/(k_1-k_2)\} - \delta n_{P_1}\{k_1k_2/(k_1-k_2)\}\left\{1/\left(D_{I2}-D_{P_1}\right)q^2 + k_2\right\} - \delta n_{P_2}\{k_1k_2/(k_1-k_2)\}\right. \\ & exp\{\left(-D_{I2}q^2 + k_2\right)t\} + \delta n_{P_1}\{k_1k_2/(k_1-k_2)\} \\ & \left[\left\{1/\left(D_{I2}-D_{P_1}\right)q^2 + k_2\right\} - \left\{1/\left(D_{I1}-D_{P_1}\right)q^2 + k_1\right\}\right]exp\left(-D_{P_1}q^2 t\right) + \delta n_{P_2} \\ & \{k_1k_2/(k_1-k_2)\}\left[\left\{1/\left(D_{I2}-D_{P_2}\right)q^2 + k_2\right\} - \left\{1/\left(D_{I1}-D_{P_2}\right)q^2 + k_1\right\}\right]exp\left(-D_{P_2}q^2 t\right)\end{aligned}$$

(eq. S1)

where δn_i and D_i (i = R, I_1, I_2, P_1, P_2) are the refractive index changes and diffusion coefficients of species i, respectively.

SI-4. Dynamic light scattering (DLS) signal of T7 RNAP

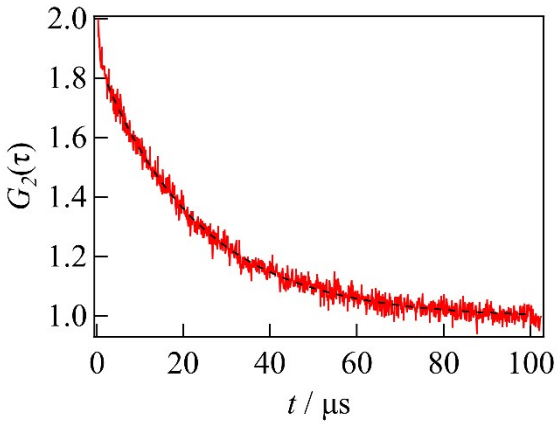

Figure S3. Autocorrelation function recorded by the dynamic light scattering for T7 RNAP at a scattering angle of 90 degree. The dotted line represents the fitting curve by a single exponential function.