
Brownian spin-locking effect

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Section S1. Observation of the Brownian spin-locking effect (BSLE) from opposite directions

As shown in Fig. S1, the spin distribution (xy plane) from a single AuNP is a four-lobe pattern. When the observation branch is along the $+x$ direction (as we did in the main text), s_ρ has the same sign as that of s_x , which is defined by $(I_R - I_L)/(I_R + I_L)$ from $+x$ direction. Here, I_R and I_L are the intensities of right- and left-handed circular polarization that are detected by inserting a quarter-wave plate and a linear polarizer between the sample and the camera. Note that this definition of handedness is associated with the propagation direction of light ($\mathbf{k}$), so that the spin direction of the electric field for I_R or I_L is opposite for the $+x$ and $-x$ propagation light. This leads to an interesting result depicted in Fig. S1: when we observe the scattering from an opposite direction, the spin distribution is flipped. The reddish color in the experimental images represents right-circular polarization, while the blueish color is for left-circular polarization (Fig. S1). The experimental demonstration is described below. We used a similar experimental setup as depicted in Fig. 1a. A linear polarizer is used between the laser ($\lambda = 639$ nm) and the AuNPs sample to generate x -polarized incident light propagating along the z direction. We symmetrically set two observation branches to detect spin of scattered field on $+x$ direction and $-x$ direction. The lenses enable cameras to receive the images of AuNPs by focusing. The cameras captured a series of images at a frame rate of 100 ms and the exposure time is 30 ms.

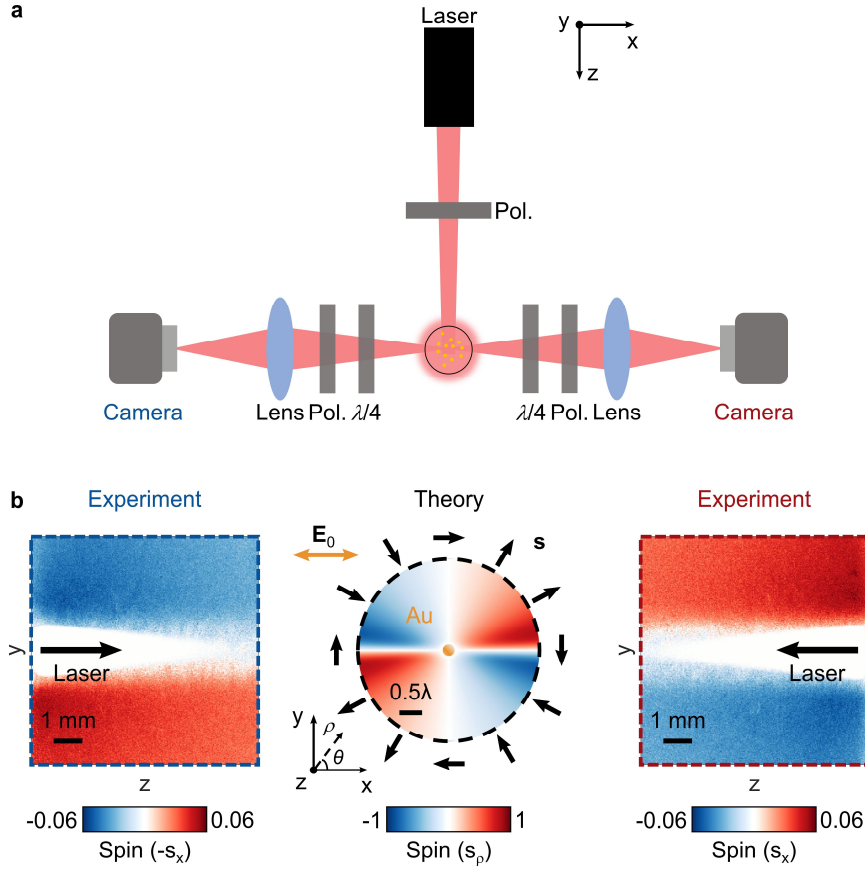


Fig. S1 Experimental observation of the BSLE of light scattered from nanoparticles. (a) Schematic of the optical setup. Laser, $\lambda = 639 \text{ nm}$; Pol., linear polarizer; $\lambda/4$, quarter-wave plate; Lens, focal length is 80 mm. (b) Middle: the four-lobe spin (s_p) distribution of the first scattering field from an AuNP ($D = 150 \text{ nm}$). s_p represents the radial component of SAM density in the xy plane. The incident direction of laser is along $+z$ direction. $\mathbf{E}_0$ represents the incident polarization. The black arrows are the normalized $\mathbf{s}$. Left and right: Experimentally observed spin-resolved images from $-x$ direction (left) and $+x$ direction (right). The reddish color stands for right-circular polarization, and the bluish color for left-circular polarization.

Section S2. Comparison of BSLE under different beam waists and polarizations

There are two key points to clarify in our BSLE. (1) This effect arises from the scattering of many nanoparticles, which is distinguished from the transverse spin effect of the incident laser beam. (2) It is worthy to demonstrate the behavior of the effect for circular polarization illumination. For these purposes, we conducted additional experiments in this section.

To show that our effect is independent from the transverse spin of the incident beam, we performed two groups of experiments, one is set in consistent with our main text setup using a plane wave (Gaussian beam waist, 2 mm), and the other uses a focused beam (focused beam waist $< 0.5 \text{ mm}$). The experimental results are shown in Fig. S2a and 2b for two different polarization states. The spin distributions for plane wave and focused

beam illumination are very similar. Therefore, the spin effect is not impacted by the beam waist of incident laser. We also provide a summarized theory for the transverse spin of focused beam in Section S9, where we can see many different properties from our BSLE compared to the transverse spin of a focused laser. Additionally, we used opposite circular polarizations as incident excitations to observe the phenomena (Fig. S2 c, d). In this case, the illumination region shows stronger spin values, while the diffusion region weaker spin values.

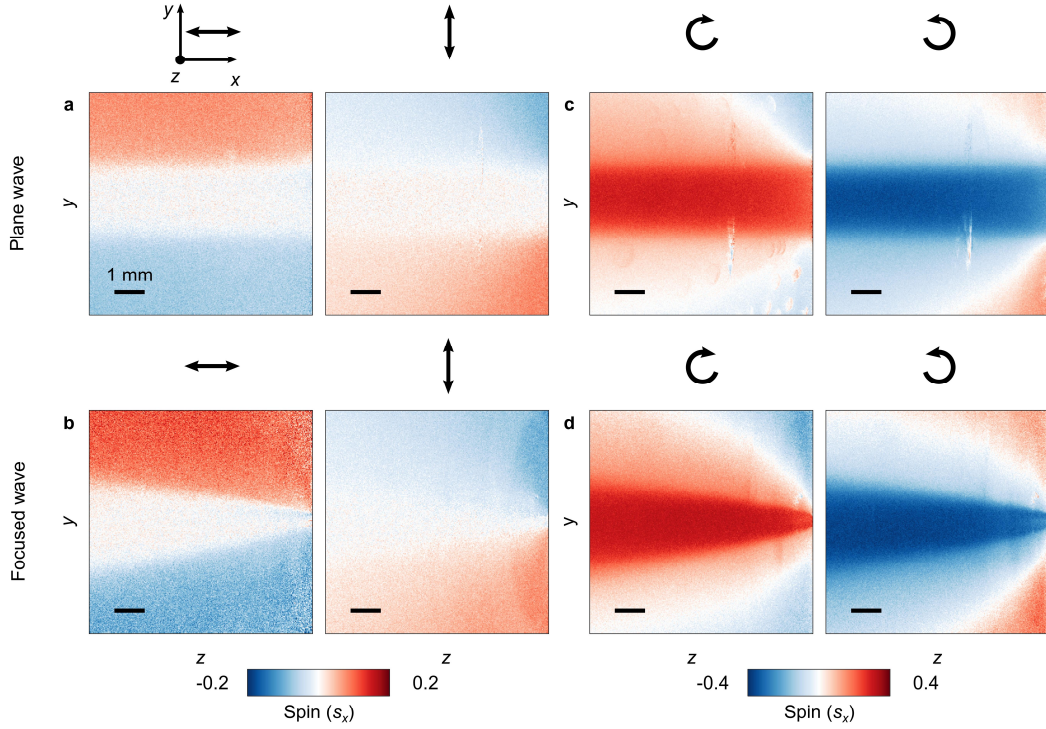


Fig.S2 Observations under different beam waists and circular polarizations. From left to right, x polarization, y polarization, left-handed circular polarization and right-handed circular polarization. The incident light is a plane wave in the first row, and a focused beam in the second row.

Section S3. Experimental setup for the spin-resolved spectra measurements

Here we show the experimental details for the observation of main text Figure 5. We used a supercontinuum laser (YSL-photonics, SC-PRO-M) to illuminate the AuNPs and analyzed the spin resolved spectra at the same diffusion region (Fig. S3). The setup is illustrated in Fig. S3. A pinhole is placed on the imaging plane to locate a region in the diffusion part for spectrum analysis. It should be noted that the spin distribution is almost homogeneous in each diffusion region, so it is easy to find a location away from the illumination region for detection. After the pinhole is set, we used a spectrometer (Zolix Omni- λ 5028i) to detect $I_R(\lambda)$ and $I_L(\lambda)$.

Afterwards, $s_x(\lambda)$ is obtained from $[I_R(\lambda) - I_L(\lambda)]/[I_R(\lambda) + I_L(\lambda)]$.

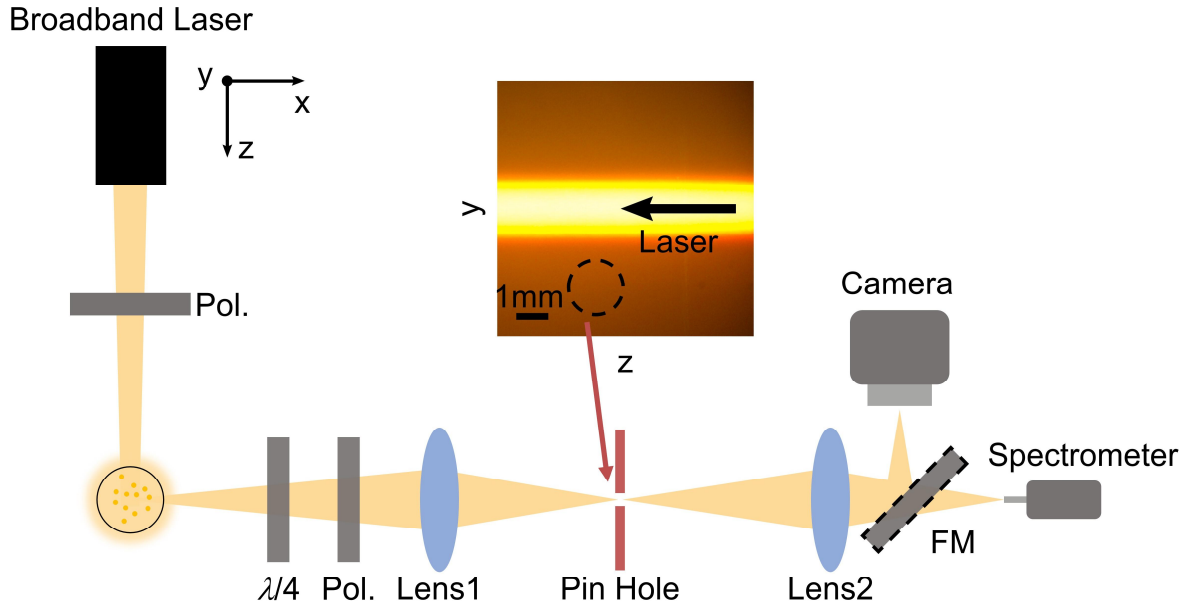


Fig.S3 Schematic of experimental setup for spin-resolved spectroscopy. We used a size-adjustable pinhole to select a small region (in the diffusion region) on the imaging plane to analyze the spin-resolved spectra (dashed circular region). The spectrometer (Zolix Omni-λ5028i) was used to capture the spin-resolved scattering spectrum of AuNPs. Broadband laser, YSL-photonics, SC-PRO-M; Pol., linear polarizer; λ/4, quarter-wave plate; FM, flip mirror; Lenses (Lens1 and Lens2, with a focal length of 80 mm).

Section S4. Observed BSLE under different linear polarizations

This section provides detailed experimental results under different incident polarizations, as an extension of main text Fig. 4. The spin images are shown in Fig. S4.

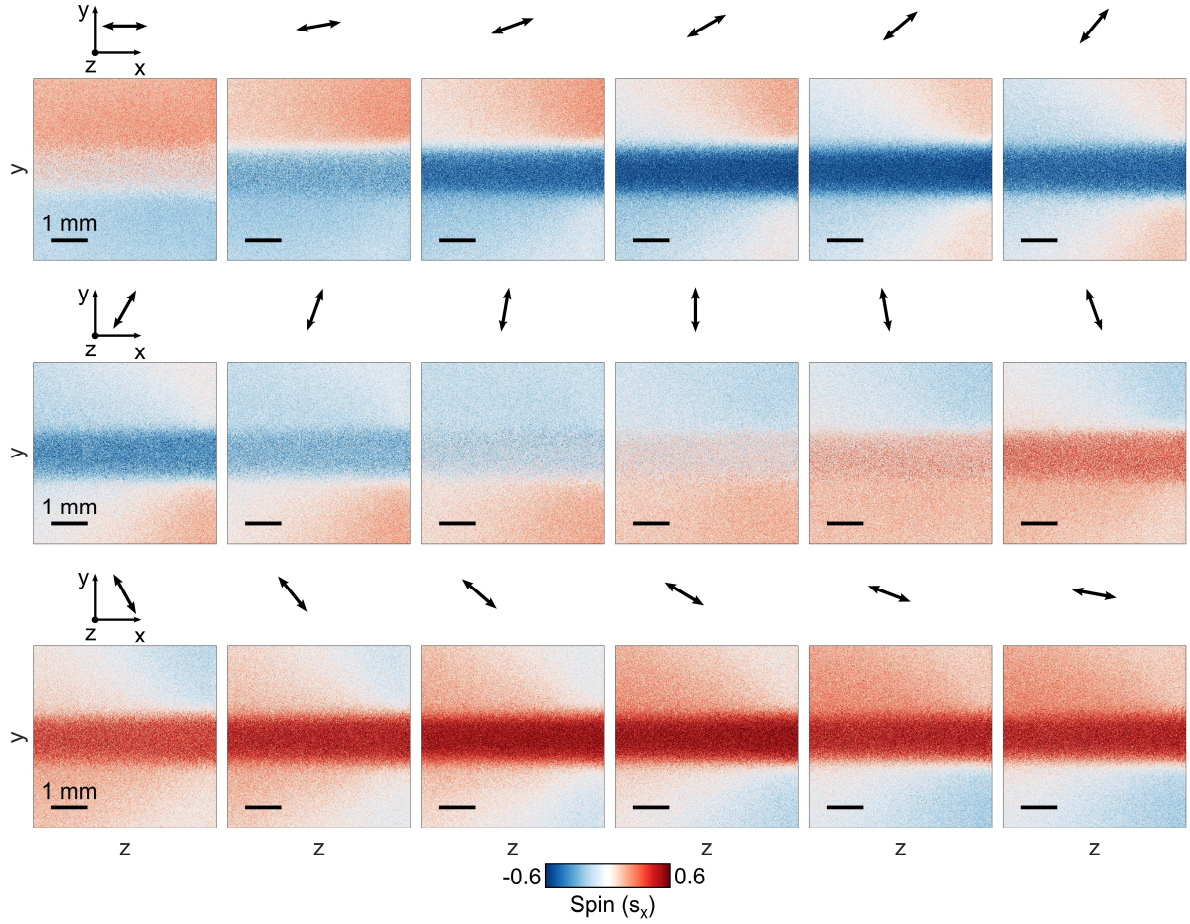


Fig.S4 Detailed experimental results of the BSLE under different incident linear polarizations. The black arrows on the top of each image represent the polarization of incident light in the xy plane. The images are captured in the yz plane, for the sample of AuNPs $\langle D \rangle = 200$ nm.

Section S5. Observed BSLE from different concentrations of nanoparticles

Our main text scattering theory has simplified the multiple scattering process into two steps. This is essential for theoretical prediction of the BSLE from an enormous number of nanoparticle scattering. In reality, complex multiple scattering will occur to affect the results, which is described by a depolarization process (1, 2). Here, we experimentally show the behavior of BSLE from nanoparticles at different concentrations (defined by the average number of nanoparticles in a unit volume). The experimental results show that even for high concentrations, the BSLE still survives, indicating its robustness against multiple dynamical scattering (Fig. S5). The details of experiments are presented below. The setup is the same as that in Fig. 1a, and the incident light is x -polarized. The tested samples are Fe_3O_4 nanoparticles (Fig. S16) with concentrations ranging from $0.57 \times 10^8 \text{ cm}^{-3}$ to $5.70 \times 10^8 \text{ cm}^{-3}$. As shown in Fig. S5a, the illumination region becomes weaker as the concentration increases. The scattering is completely diffusive within the sample, as the concentration reaches $5.70 \times 10^8 \text{ cm}^{-3}$. This means that multiple scattering, rather than single or few scatterings, has dominated. The

spin images are presented during this process, as shown in Fig. S5b. It can be seen that the BSLE is strong for small concentrations. As the concentration increases, the spin effect becomes weaker, although its spatial distribution remains similar. Specifically, at longer propagation distances, the spin decreases faster. We particularly take two points in the image for analysis, ① and ②, which stand for longer (~ 1 cm) and shorter (~ 1 mm) propagation distances in the scattering medium, and their intensities and spin evolution as a function of concentration are depicted in Fig. S5c and d, respectively. While the intensity evolution in both regions (① and ②) is comparable across the concentration range, the spin distributions differ significantly. Specifically, when the concentration is approximately $2.28 \times 10^8 \text{ cm}^{-3}$, the statistically averaged spin at region ①, $\langle s_{x1} \rangle$, approaches 0, while $\langle s_{x2} \rangle$ is about 0.09. Notably, the s_x distribution near region ② persists over a wide range of concentrations, indicating the robustness of the BLSE across single and multiple scattering regimes.

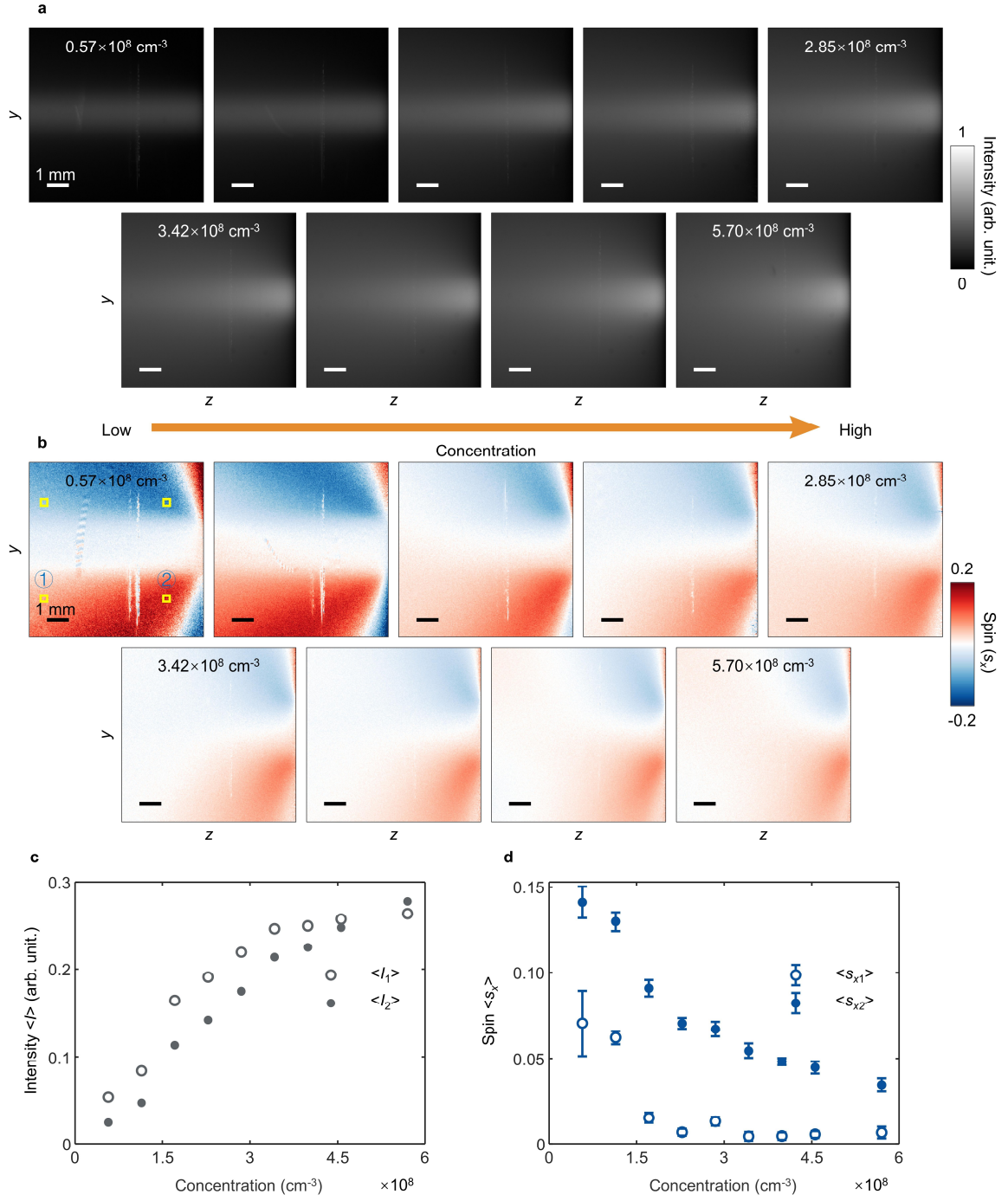


Fig.S5 Experimentally observed Brownian spin-locking effect at different Fe₃O₄ nanoparticle concentrations. Normalized light intensity distributions **(a)** and s_x distributions **(b)** for various concentrations of Fe₃O₄ nanoparticles in water. Yellow boxes ① and ② denote the analysis regions used to extract the light intensity and s_x values in **(c)** and **(d)**. The two upper yellow boxes represent regions symmetric to ① and ② with respect to the laser beam. The laser is incident from the right side of the images. **(c)** Variation of normalized light intensity with particle concentration. $\langle I_1 \rangle$ ($\langle I_2 \rangle$) represents the average light intensity in region ① (②). **(d)** Variation of $\langle s_x \rangle$ with particle concentration. $\langle s_{x1} \rangle$ ($\langle s_{x2} \rangle$) represents the average absolute value of s_x in region ① (②). Data are presented as mean $\pm$ s.d. (technical replicates, $n = 10$).

Section S6. Extracting the value of spin at different detector exposure times

To confirm that the experimental value of s_x is accurate, we changed the exposure time of the camera and analyzed the evolution of s_x to find a suitable exposure time and incident power. The experimental setup is same as that in Fig. 1a and the incident light is x -polarized. The power of incident laser is set to a moderate value of about 5 mW (continuous wave). The exposure time of the camera can be set from 0.1 ms to 12 ms. The intensities are shown in Fig. S6a, where we see the intensity increases with the exposure time. On the one hand, a sufficiently long exposure time allows us to observe a stable spin of the scattered light over a period within the Brownian motion nanoparticle system, as shown in Fig. S6b. On the other hand, we need to avoid light intensity saturation, which also distorts the value of s_x . As shown in Fig. S6c and d, when the exposure time is longer than 1 ms, $\langle s_x \rangle$ is approximately stable in the diffusion region. This gives us a guideline to set the camera properly in order to obtain correct s_x . All experimental results are achieved after we perform this exposure time adjustment.

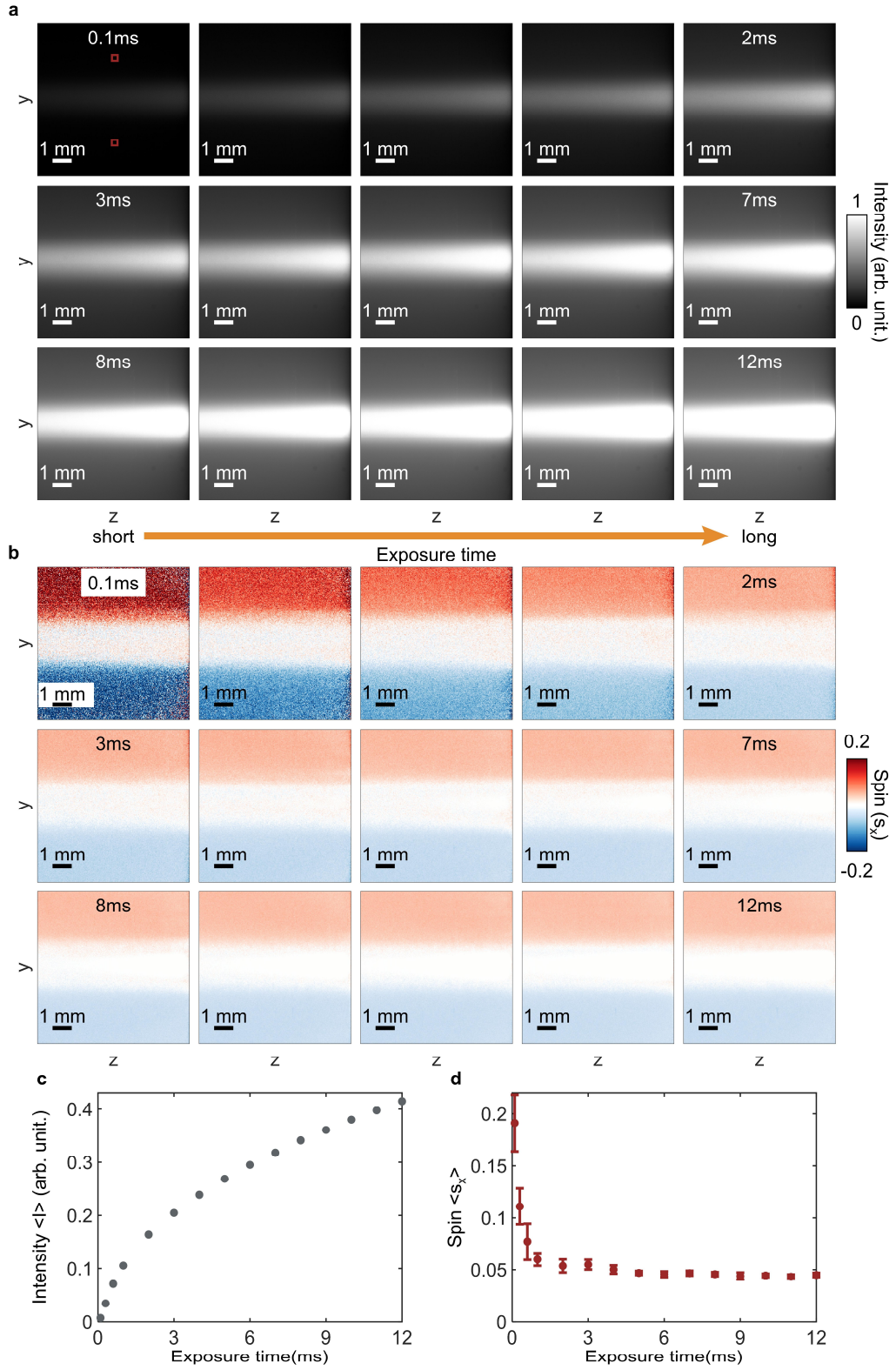


Fig.S6 Captured s_x and light intensity at different exposure times of camera. Normalized light intensity distribution **(a)** and s_x distribution **(b)** from different exposure times of camera. The red boxes in **(a)** represent the analysis regions to obtain light intensity and s_x information for **(c)** and **(d)**. **(c)** The change in average light intensity in the selected region with respect to exposure time. **(d)** The change in $\langle s_x \rangle$ with respect to exposure time. $\langle s_x \rangle$ represents the average absolute value of s_x in the two red box regions. Data are presented as mean $\pm$ s.d. (technical replicates, $n = 25$).