Supplementary Information

Rapid-Prototyping a Brownian Particle in an Active Bath

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Theoretical overview

The tracer particle is kicked by a series of active forces, each of which is generated randomly at the time t_i with random amplitude. Each kick shifts the center of the trap decaying exponentially with characteristic time τ_c, which is schematically shown in Fig. 1. Then, the active force acting on the tracer particle at a certain time t is the sum of active forces remaining until the time t, given as

$$\xi_{act}(t) = k\sum_i d_i \mathrm{e}^{-(t-t_i)/\tau_c} \cdot \theta(t-t_i) = kx_c(t),$$ * MERGEFORMAT (S1)

where $\theta(t-t_i)$ is the step function equal to 1 for $t > t_i$ and 0 otherwise. d_i is a Gaussian random number with variance X^2 and t_i is produced by the Poisson distribution with mean interval τ_P. The resultant position $x_c(t)$ of the center of the shifted potential is equal to the sum of the forces divided by k, given in the above equation.

The particle position can be expressed as $x \equiv x_{th} + x_{act}$, where x_{th} is due to the thermal noise and x_{act} due to the active force. Therefore, Eq. (1) in the text can be expressed as a set of two stochastic differential equations which are completely independent to each other.

$$\gamma \dot{x}_{th} = -kx_{th} + \xi_{th}, \qquad \gamma \dot{x}_{act} = -kx_{act} + \xi_{act}$$ * MERGEFORMAT (S2)

Here, $\xi_{th}(t)$ is a usual thermal noise with zero mean and time-correlation $\langle \xi_{th}(t)\xi_{th}(t') \rangle = 2\gamma k_B T \delta(t-t')$ and $\xi_{act}(t)$ is an active force in Eq. (S1). The time-correlation function of active forces is useful to investigate the stochastic properties of the system such as $\langle x^2 \rangle$, $\langle x(t)x(t') \rangle$ etc., where $\langle \cdots \rangle$ denotes the average over thermal and active forces. We find for $t' > t$

$$\left\langle \xi_{act}(t)\xi_{act}(t')\right\rangle = k^2 X^2 \mathrm{e}^{-(t'-t)/\tau_c}\left\langle \sum_i \mathrm{e}^{-2(t-t_i)/\tau_c}\right\rangle,$$ * MERGEFORMAT

(S3)

where we use $\langle d_i d_j\rangle = X^2\delta_{ij}$. Using the property of the Poisson distribution,

$$\left\langle \sum_i \mathrm{e}^{-2(t-t')/\tau_c}\right\rangle = \int_0^t \frac{ds}{\tau_p}\mathrm{e}^{-2(t-s)/\tau_c}.$$ * MERGEFORMAT (S4)

Using Eq. (S4) for steady-state condition ($t >> \tau_c$), the noise auto-correlation function in Eq. (S3) takes the following form

$$\left\langle \xi_{act}(t)\xi_{act}(t')\right\rangle = k^2 X^2 \frac{\tau_c}{2\tau_P}\mathrm{e}^{-(t'-t)/\tau_c}.$$ * MERGEFORMAT (S5)

The exponential correlation in this equation is similar to that for the OU noise, but higher-order cumulants are present for all orders, unlike the OU noise. As τ_c/τ_p increases, the active noise becomes the OU noise, as seen in Fig. 1(c) in the text. We can observe such a tendency as the kurtosis $\propto \tau_p/\tau_c$ decreases.

One of the Referees provided a rigorous proof for the transition to the OU noise in the reviewing process. We will summarize the proof in the following. Integrating Eq. (3) from t to $t+dt$, $\xi_{act}(t+dt)=\xi$ and $\xi_{act}(t)=\xi'$ are related as $\xi = \xi' - \xi'/\tau_c dt + h$, where $h = kd$ for random amplitude d with probability dt/τ_p and $h=0$ with probability $1-dt/\tau_p$. Then, the Kolmogorov equation for Eq. (3) is written up to the first order in dt as

$$P(\xi,t+dt)=\int d\xi' P(\xi',t)[\delta\left(\xi-\xi'+\xi'/\tau_c dt\right)\left(1-dt/\tau_p\right)$$
$$+\delta\left(\xi-\xi'+\xi'/\tau_c dt-hd\right)\left(dt/\tau_p\right)]$$
* MERGEFORMAT
$$=P(\xi,t)+\frac{dt}{\tau_p}\left[P(\xi-ky)-P(\xi)\right]+\frac{dt}{\tau_c}\frac{\partial}{\partial\xi}\xi P(\xi,t).$$

(S6)

Here, $\delta(\xi-\xi'+cdt)=(1-cdt\,\partial/\partial\xi')\delta(\xi-\xi')$ is used and the integration is done over ξ'.

Then, expanding $P(\xi-ky)$ in powers of y and averaging over y, we get

$$\frac{\partial P(\xi,t)}{\partial t}=\frac{1}{\tau_c}\frac{\partial}{\partial\xi}\xi P(\xi,t)+\frac{1}{\tau_p}\sum_{n=1}^{\infty}\frac{k^{2n}C_{2n}}{n!}\frac{\partial^n}{\partial\xi^n}P(\xi,t).$$ *

MERGEFORMAT (S7)

Here, $C_{2n}=\left\langle d^{2n}\right\rangle$ with the bracket denoting the average over y. By changing variables

as $T=t/\tau_c$ and $X=\xi/\sqrt{\Omega}$ for $\Omega=\tau_c/\tau_p$, we find

$$\frac{\partial P(X,T)}{\partial t}=\frac{\partial}{\partial X}XP(X,T)+\sum_{n=1}^{\infty}\frac{k^{2n}C_{2n}\Omega^{1-n}}{(2n)!}\frac{\partial^{2n}}{\partial X^{2n}}P(X,T).$$ *

MERGEFORMAT (S8)

In the limit $\Omega\to\infty$, we get the OU process

$$\frac{\partial P(X,T)}{\partial t}=\frac{\partial}{\partial X}XP(X,T)+\frac{k^2C_2}{2}\frac{\partial^2}{\partial X^2}P(X,T).$$ * MERGEFORMAT

(S9)

We initially prepared the system in equilibrium in the absence of the active force. Then, the PDF of x_{th} remains Boltzmann every time, and hence that of $x-x_{act}(t)$ for a given $x_{act}(t)$ such that

$$P\left(x,t\,|\,x_{act}\right)=\sqrt{\frac{k}{2\pi k_BT}}\,\mathrm{e}^{-k\left(x-x_{act}(t)\right)^2/2k_BT} \quad .$$ * MERGEFORMAT

(S10)

This conditional PDF of x for $x_{act}(t)$ is Gaussian, while the PDF averaged over $x_{act}(t)$

becomes non-Gaussian in general.

From Eq. (S2), we have

$$x_{th}(t)=\frac{1}{\gamma}\int_0^t ds\mathrm{e}^{-(t-s)/\tau_k}\xi_{th}(s),\quad x_{act}(t)=\frac{1}{\gamma}\int_0^t ds\mathrm{e}^{-(t-s)/\tau_k}\xi_{act}(s).$$ *

MERGEFORMAT (S11)

The variance of each position is calculated from Eqs. (S5) and (S11), given as

$$\left\langle x_{th}^2\right\rangle=\frac{k_BT}{k},\quad \left\langle x_{act}^2\right\rangle=\frac{X^2}{2}\frac{\tau_c}{\tau_p}\left(1+\frac{\tau_k}{\tau_c}\right)^{-1}=\frac{k_BT_{act}}{k},$$ *

MERGEFORMAT (S12)

where T_{act} is tracer activity which increases due to active force and the long-time limit t ? τ_c is used. Then, we find the variance of $x=x_{th}+x_{act}$ as

$$\left\langle x^2\right\rangle=\left\langle x_{th}^2\right\rangle+\left\langle x_{act}^2\right\rangle=\frac{k_B\left(T+T_{act}\right)}{k}.$$ * MERGEFORMAT

(S13)

The full statistics requires the information of all the moments of higher orders due to a non-Gaussian property of x_{act}, which is practically not possible. Fig. S1 shows the PDFs of x, x_{act}, and x_{th}. For large τ_c/τ_p, $P(x_{act})$ and $P(x)$ are close to Gaussian distributions because a large number of peaks in correlation time τ_c behave as Gaussian due to the central limit theorem.

***Van Hove self-correlation* function $G_s(\Delta x,\Delta t)$**

It is defined as the PDF of the particle displacement Δx during Δt. We estimate the non-Gaussian parameter given in the text as $\alpha_2 \equiv [< x^4 > /5 < x^2 >^2] - 3/5$. The result is shown in Fig. S2.

Mean square displacement (MSD) of a free particle in an active bath

If we consider a free particle in an active bath (without harmonic potential), the Langevin equation is described as $\gamma \dot{x}(t) = \xi_{th}(t) + \xi_{act}(t)$. Then the position of the particle $x(t)$ is expressed as

$$\Delta x(t) = \frac{1}{\gamma}\int_0^t dt' \left[\xi_{th}(t') + \xi_{act}(t')\right],$$ * MERGEFORMAT

(S14)

and the mean square displacement $\left\langle \Delta x^2(t) \right\rangle$ of the particle is

$$\left\langle \Delta x^2(t) \right\rangle = \frac{1}{\gamma^2}\int_0^t dt' \int_0^t dt'' \left\langle \xi_{th}(t')\xi_{th}(t'') \right\rangle + \frac{1}{\gamma^2}\int_0^t dt' \int_0^t dt'' \left\langle \xi_{act}(t')\xi_{act}(t'') \right\rangle.$$ *

MERGEFORMAT (S15)

Using $\left\langle \xi_{th}(t')\xi_{th}(t'') \right\rangle = 2\gamma k_B T \delta(t' - t'')$, $\left\langle \xi_{act}(t')\xi_{act}(t'') \right\rangle \equiv C \mathrm{e}^{-\left|\frac{t'-t''}{\tau_c}\right|}$, and $C = \gamma^2 \frac{D_{act}}{\tau_c} = \frac{f_{RMS}^2 \tau_c}{2\tau_p}$, we can simplify Eq. (S11). Here the f_{RMS} is defined as random active kick force. (In harmonic potential, the active force satisfies this relationship; $f_{RMS} = kX$)

$$\left\langle \Delta x^2(t) \right\rangle = 2\frac{k_B T}{\gamma} t + \frac{2C\tau_c}{\gamma^2}\int_0^t dt' \mathrm{e}^{-t'/\tau_c}\left(\mathrm{e}^{t'/\tau_c} - 1\right)$$ *

MERGEFORMAT (S16)

Finally, the MSD of a free particle in an active bath is expressed as

$$\left\langle \Delta x^2(t) \right\rangle = 2D_{th}t + 2D_{act}\left[t - \tau_c \left(1 - e^{-t/\tau_c}\right)\right].$$ MERGEFORMAT * (S17)

Power spectral density

We can apply the Fourier transformation for the position and noise variables in a steady-state where we can neglect initial memory terms containing e^{-t/τ_k}. We write $x(t) = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} d\omega e^{i\omega t} x(\omega)$ and $x(\omega) = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} d\omega e^{-i\omega t} x(t)$. Then, Eq. (S2) yields

$$x(\omega) = \frac{\xi_{th}(\omega) + \xi_{act}(\omega)}{k + i\gamma\omega}.$$ MERGEFORMAT * (S18)

Using $\left\langle \xi_{th}(t)\xi_{th}(t') \right\rangle = 2\gamma^2 D_{th}\delta(t-t')$ and $\left\langle \xi_{act}(t)\xi_{act}(t') \right\rangle = \gamma^2 D_{act}\cdot\left(1+\tau_c/\tau_k\right)\cdot e^{-|t-t'|/\tau_c}/\tau_c$ from Eq. (S3), we get

$$\begin{aligned}\left\langle \xi_{th}(\omega)\xi_{th}(\omega') \right\rangle &= 2\gamma^2 D_{th}\delta(\omega+\omega'),\\ \left\langle \xi_{act}(\omega)\xi_{act}(\omega') \right\rangle &= \frac{2\gamma^2 D_{act}\cdot\left(1+\tau_c/\tau_k\right)}{1+\tau_c^2\omega^2}\delta(\omega+\omega'),\end{aligned}$$ * MERGEFORMAT (S19)

where $D_{th} = k_B T/\gamma$ (diffusion constant of the medium) and $D_{act} = k_B T_{act}/\gamma$ (active diffusion coefficient). As a result, we get $\left\langle x(\omega)x(\omega') \right\rangle = S_{xx}(\omega)\delta(\omega+\omega')$ so that

$$S_{xx}(t-t') = \left\langle x(t)x(t') \right\rangle = \int_{-\infty}^{\infty}\frac{d\omega}{2\pi}C_{xx}(\omega)e^{i\omega(t-t')}.$$ * MERGEFORMAT (S20)

Using Eq. (S18), the correlation function (spectral density) of position in frequency space is given as

$$S_{xx}(\omega)=\frac{2/\tau_k}{\omega^2+\left(1/\tau_k\right)^2}\left[\frac{k_B T}{k}+\frac{k_B T_{act}}{k}\cdot\left(1+\frac{\tau_c}{\tau_k}\right)\cdot\frac{1}{1+\omega^2\tau_c^2}\right].$$ *

MERGEFORMAT (S21)

It agrees well with the experimental results shown in Fig. S3.

Heat dissipation

The violation of fluctuation-dissipation relation

In nonequilibrium process, incessant heat production Q is found to be related to the violation of the fluctuation-dissipation theorem. Let $C(t,t')=\left\langle v(t)v(t')\right\rangle$ and $R(t,t')$ be correlation function and response functions for velocity, respectively. The violation of the fluctuation-dissipation relation (FDR) is given for $t>t'$ as

$$C(t,t')-2k_B TR(t,t')=\frac{1}{2\gamma}[\left\langle v(t)\left(-kx(t')+\xi_{act}(t')\right)\right\rangle$$
$$+\left\langle v(t')\left(-kx(t)+\xi_{act}(t)\right)\right\rangle].$$ *

MERGEFORMAT (S22)

The right side in this equation becomes $\gamma^{-1}\left\langle\dot{Q}\right\rangle$ in the limit $t\to t'$, where $\left\langle\dot{Q}\right\rangle=\left\langle\dot{x}(t)\left(\gamma\dot{x}(t)-\xi_{th}(t)\right)\right\rangle$ is the average rate of heat dissipation. To obtain the response function, an infinitesimal perturbation $f^p(t)$ is applied to the system. In this case, the Langevin equation is written by

$$\gamma\dot{x}(t)=-kx(t)+\xi_{th}(t)+\xi_{act}(t)+f^p(t).$$ * MERGEFORMAT

(S23)

The general solution of Eq. (S23) is $x(t)=\gamma^{-1}\int_0^t dt' \mathrm{e}^{-(t-t')/\tau_k}\left(\xi_{th}(t')+\xi_{act}(t')+f^p(t')\right)$, where the initial memory term $x(0)\mathrm{e}^{-t/\tau_k}$ is neglected in a steady-state. The ensemble average of the velocity is $\langle v(t)\rangle=\frac{1}{\gamma}\left[f^p(t)-\frac{1}{\tau_k}\int_0^t dt' \mathrm{e}^{-(t-t')/\tau_k} f^p(t')\right]$ and the response function $\delta\langle v(t)\rangle/\delta f^p(t')$ is given as

$$\chi(t,t')=\frac{1}{\gamma}\left[\delta(t-t')-\frac{1}{\tau_k}\mathrm{e}^{-(t-t')/\tau_k}\theta(t-t')\right].$$ MERGEFORMAT

(S24)

Note that the response function is independent of active force as well as thermal force. Fig. S4A shows the active force and perturbation force at the time $t=t_0$. Fig. S5B shows that the presence of the active force $\xi_{act}(t)$ on the system does not change the response time τ_R. In this experiment the response time is $\tau_R=\tau_k=1.1\,m\mathrm{s}$.

The average rate of heat dissipation in a steady state

The velocity correlation function in frequency space is equal to $C(\omega)=\langle v(\omega)v(-\omega)\rangle=\omega^2 S_{xx}(\omega)$. Also, the amplitude of response function can be calculated by Eq. (S23).

$$R(\omega)\equiv \mathrm{Re}\left(\chi(\omega)\right)=\frac{1}{\gamma}\cdot\frac{\omega^2}{\omega^2+1/\tau_k^2}$$ MERGEFORMAT

(S25)

Then, the steady-state(t,t' ? τ_k, $C(t,t')=C(t-t')$, and $R(t,t')=R(t-t')$) rate of heat dissipation is given as

$$\left\langle \dot{Q} \right\rangle = \gamma \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \left[C(\omega) - 2k_B T R(\omega) \right].$$ * MERGEFORMAT (S26)

We write $D(\omega) \equiv \gamma \left(C(\omega) - 2k_B T R(\omega) \right)$. From Eqs. (S21) and (S25), we get

$$D(\omega) = \left(\frac{2k_B T_{act}}{\tau_c^2} \right) \cdot \left(1 + \frac{\tau_c}{\tau_k} \right) \cdot \left[\frac{\omega^2}{\left(\omega^2 + 1/\tau_k^2\right)\left(\omega^2 + 1/\tau_c^2\right)} \right].$$ *

MERGEFORMAT (S27)

Then, we find the average rate of heat dissipation in a steady-state as

$$\left\langle \dot{Q} \right\rangle = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} D(\omega) = \frac{k_B T_{act}}{\tau_c},$$ * MERGEFORMAT

(S28)

where T_{act} is defined in Eq. (S12). The work production rate can be defined in three different ways: (a) $\dot{W} = (\partial/\partial t) k \left(x - \xi_{act}(t)/k \right)^2 / 2 = -\left(x - \xi_{act}(t)/k \right) \dot{\xi}_{act}(t)$, (b) $\dot{W} = (\partial/\partial t) \left[kx^2/2 - x\xi_{act}(t) \right] = -x\dot{\xi}_{act}(t)$, and (c) $\dot{W} = \dot{x}\xi_{act}(t)$. Here, (a) is from the viewpoint of moving potential with the center at $\xi_{act}(t)/k$, (b) is defined as the summation of harmonic potential $kx^2/2$ and active potential $x\xi_{act}(t)$, and (c) is from the interpretation of $\xi_{act}(t)$ as an external time-dependent force. In all three cases, the heat dissipation rate has the same expression $\dot{x}\left(\gamma \dot{x} - \xi_{th}(t) \right)$. In the steady-state limit, all definitions of work rate lead to the same average value equal to the average heat dissipation rate in Eq. (S28). The three values become different in the transient period, which is not examined in detail in our experiment but can be calculated rigorously.

Maximum heat dissipation rate frequency

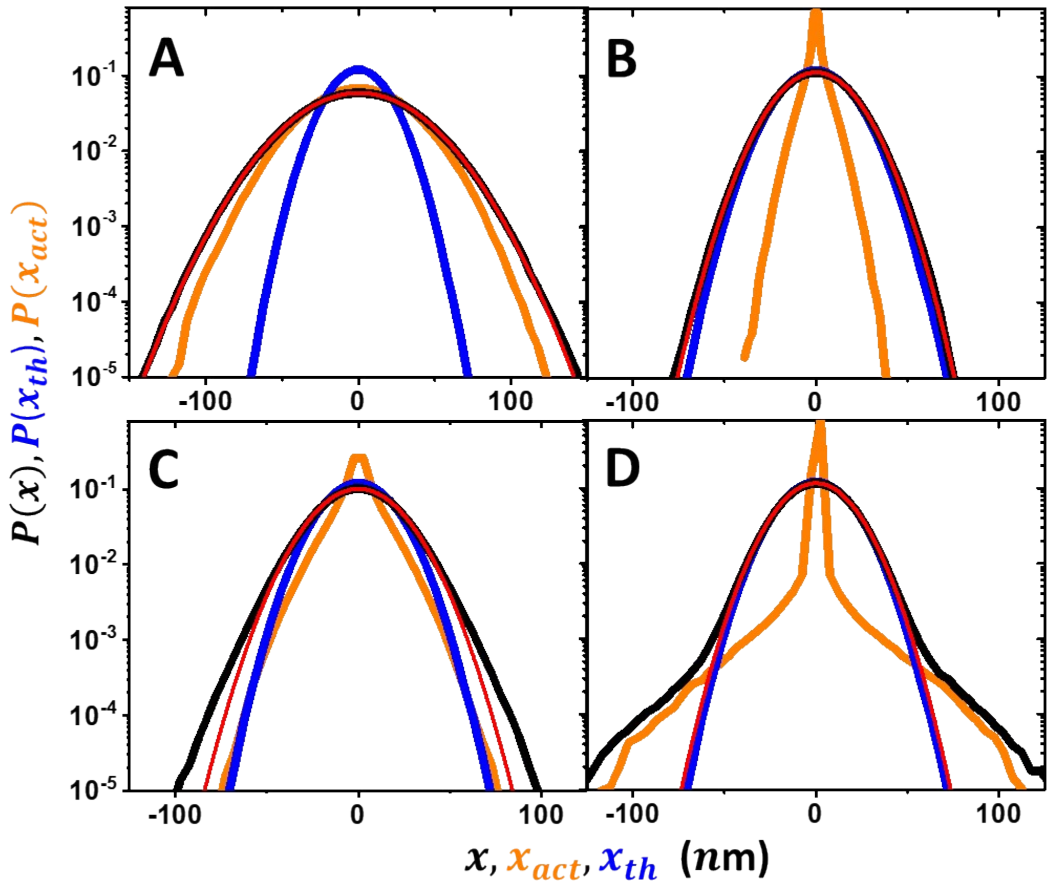
The heat dissipation rate in frequency space has the maximum value at ω_d which we get $\partial D(\omega)/\partial\omega = 0$. We find

$$\omega_d = \frac{1}{\sqrt{\tau_k \tau_c}}.$$ MERGEFORMAT

(S29)

Experiment setup

Fig. S5 shows the experimental setup in detail.



Supplementary Figures

FIG. S1. PDFs of particle position. PDFs of x (black), x_{act} (orange), and x_{th} (blue) from Eq. (1) and Eq. (S2) by using computer simulation. The red solid curves are Gaussian fittings to $P(x)$. (A) and (B) show the PDFs when $P(x)$ is Gaussian-like. (C) and (D) show the PDFs when $P(x)$ is non-Gaussian. The conditions are (A) $X = 20\,n\text{m}$, $\tau_c = 10\,ms$ and $\tau_p = 2\,ms$, (B) $X = 20\,n\text{m}$, $\tau_c = 2\,ms$ and $\tau_p = 10\,ms$, (C) $X = 40\,n\text{m}$, $\tau_c = 2\,ms$ and $\tau_p = 8\,ms$, and (D) $X = 70\,n\text{m}$, $\tau_c = 4\,ms$ and $\tau_p = 200\,ms$.

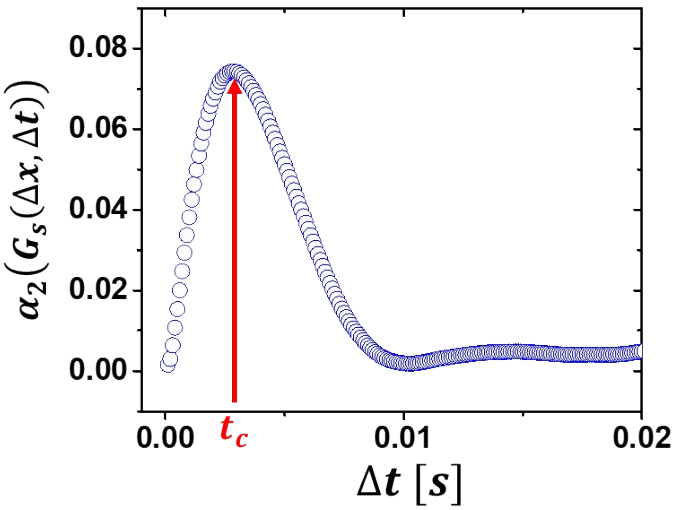


FIG. S2. The non-Gaussian parameter value of Van Hove self-correlation function $G_s(\Delta x, \Delta t)$ vs. Δt. α_2 is obtained by computer simulation for $X = 70n\text{m}$, $\tau_c = 0.3ms$, and $\tau_k = 0.8ms$. In the same condition, the non-Gaussian parameter value of $P(x)$ is 0.

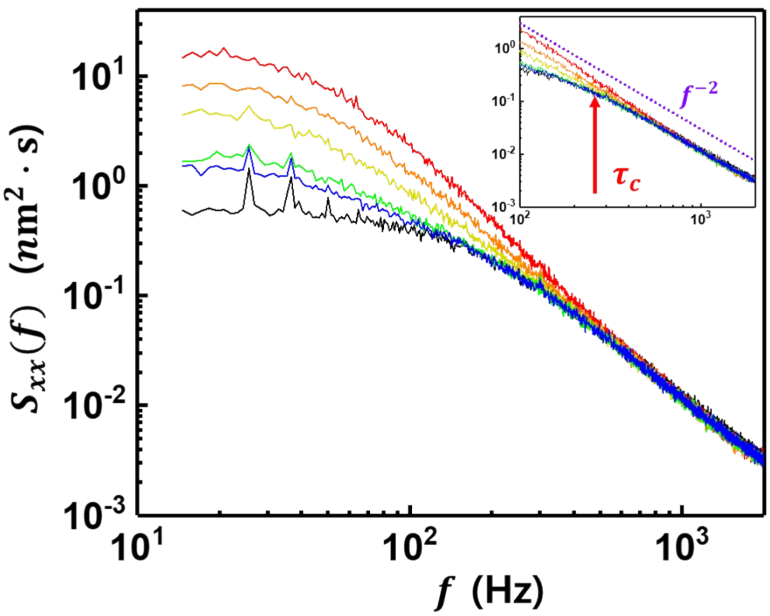


FIG. S3. Power spectral density (PSD) of particle position for various conditions; τ_c/τ_p : 4.0 (red), 2.0 (orange), 1.0 (yellow), 0.33 (green), and 0.25 (blue). Black corresponds to PSD without the active force. Here, $X = 37n\text{m}$, $\tau_c = 4m\text{s}$, and $\tau_k = 1.1m\text{s}$. In the inset, the violet dotted line indicates the slope for free diffusion.

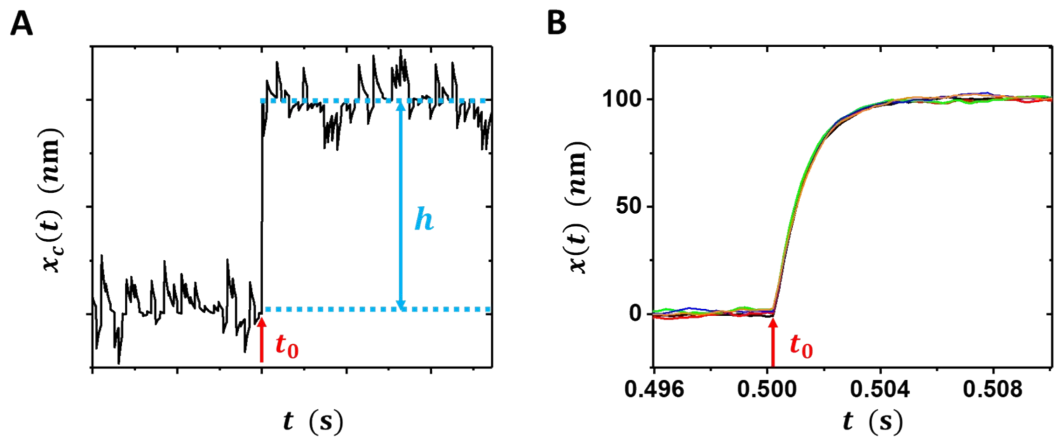


FIG. S4. Measurement of the response function. (A) The center of the trap is additionally shifted with distance h at $t = t_0$. During this process, the system is under the active force. (B) The average trajectory of the particle over 500 different measurements. The position of the particle is observed during the relaxation process in the same condition.

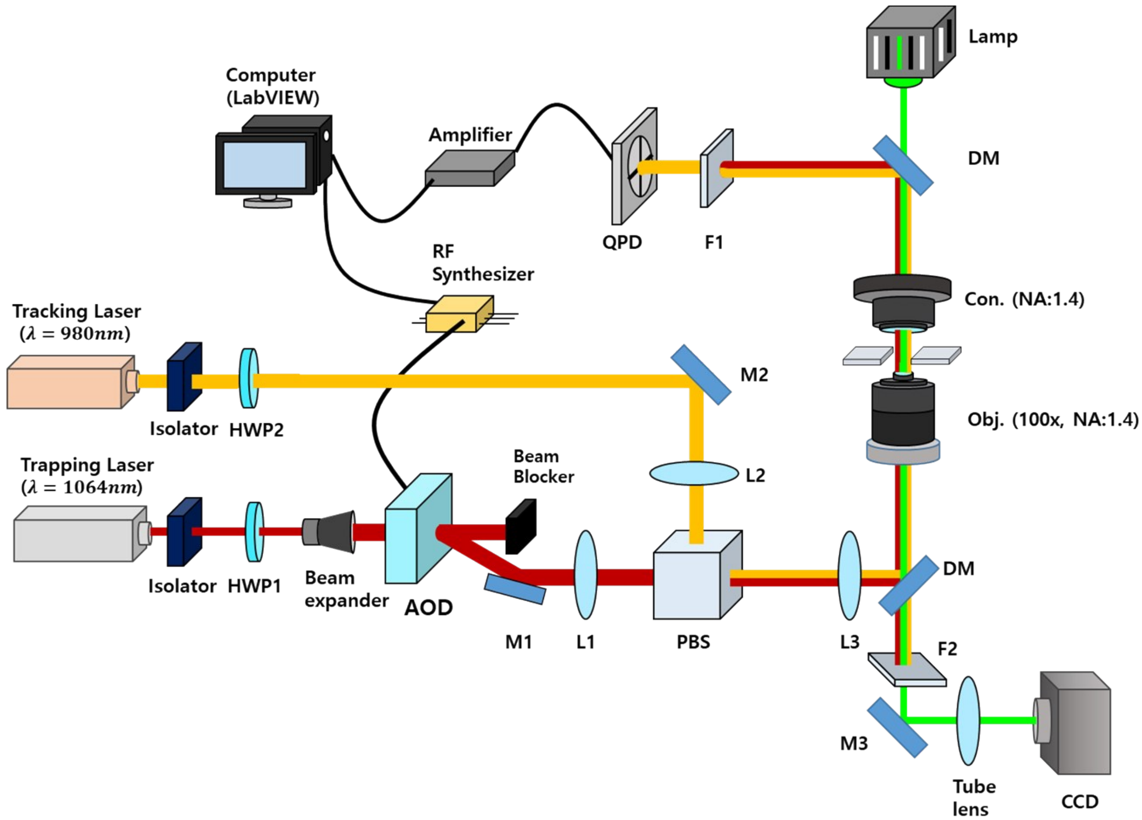


FIG. S5. Schematic diagram of experimental setup.