Electronic Supplementary Information

Estimation of effective anisotropy constant distribution of magnetic nanoparticles based on magnetic particle spectroscopy

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AC *M-H* curves obtained by MPS

The AC *M*–*H* curves of the solidified samples were measured at room temperature using magnetic particle spectroscopy (MPS) under an AC excitation field with an amplitude of $\mu_0 H_{ac} = 10\ mT$ and the frequencies ranging from $f =$ 1 to 20 kHz.

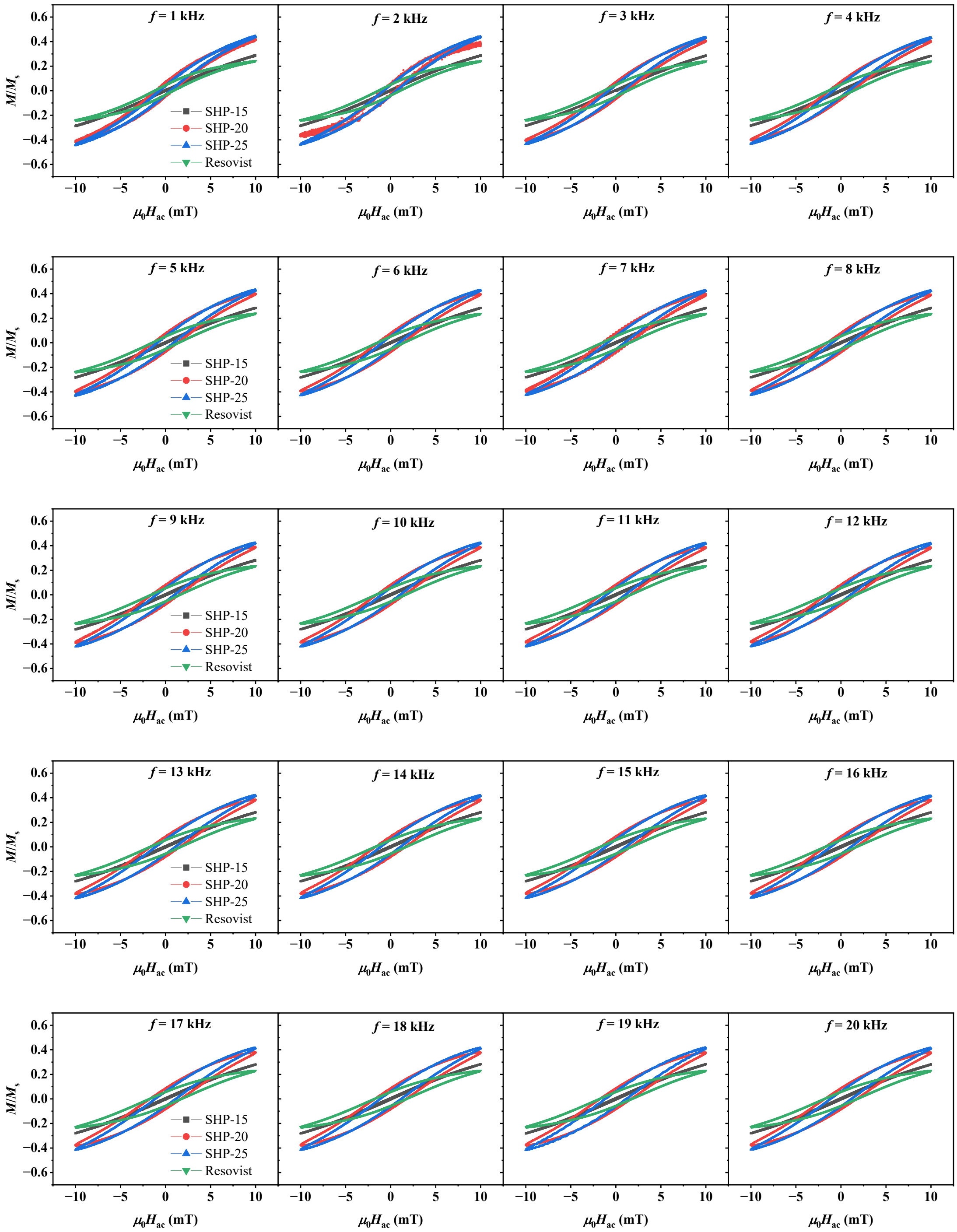

Fig. S1 AC *M-H* curves of four MNP samples.

Core size distribution estimated from static *M-H* curve obtained by VSM

The saturation magnetizations M_s of samples were measured from the static *M-H* curves using VSM. The obtained values of M_s at $T = 300\ K$ were $M_s = 353.42\ kA/m$ (SHP-15), $268.0\ kA/m$ (SHP-20), $185.48\ kA/m$ (SHP-25), $295.45\ kA/m$ (Resovist), and $282.95\ kA/m$ (MS3), respectively.
The core size distribution $n_{dc,V}(d_c)$ was estimated by analysing the static *M-H* curve. Detail cprocedures can be found in Refs [1] and [2]. Figs S2 and S3 show the core size distribution of samples. The main core diameters d_{c_main} of SHP-15, SHP-20, and SHP-25 are around 15 nm, 20 nm, and 20 nm, respectively. For Resovist, the core size distribution exhibits two main peaks: the smaller d_{c_main} around 8 nm corresponds to single core MNPs, while the larger d_{c_main} around 23 nm, represents to multi-core MNPs. For MS3, in which particles larger than 20 nm were largely removed from Resovist, d_{c_main} is approximately 8 nm.

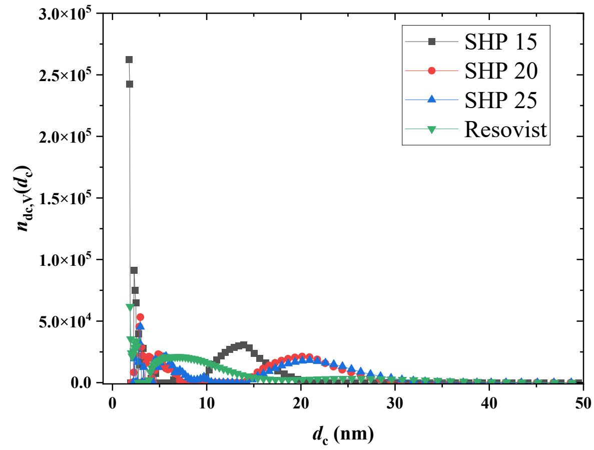

Fig. S2 Volume-weighted number density of MNPs having d_c, $n_{dc,V}(d_c)$, of SHP-15, SHP-20, SHP-25, and Resovist.

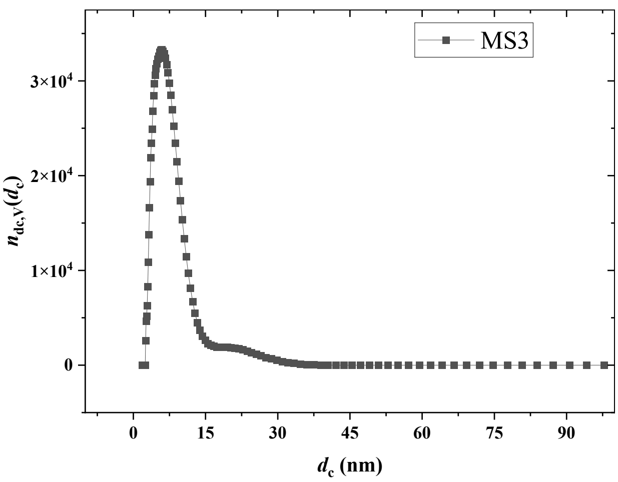

Fig. S3 Volume-weighted number density of MNPs having d_c, $n_{dc,V}(d_c)$, of MS3.

Hydrodynamic size distribution estimated by DLS

The hydrodynamic size d_h of samples were measured using dynamic light scattering (DLS). The main hydrodynamic sizes d_{h_main} are approximately 43 nm, 50 nm, 55 nm and 70 nm for SHP-15, SHP-20, SHP-25 and Resovist, respectively. The SHP samples exhibit relatively narrow and monomodal distributions, indicating uniform dispersion of nanoparticles in aqueous solution. In contrast, Resovist shows a broader and more dispersed d_h distribution which can be attributed to the presence of two d_c peak in core size distribution.

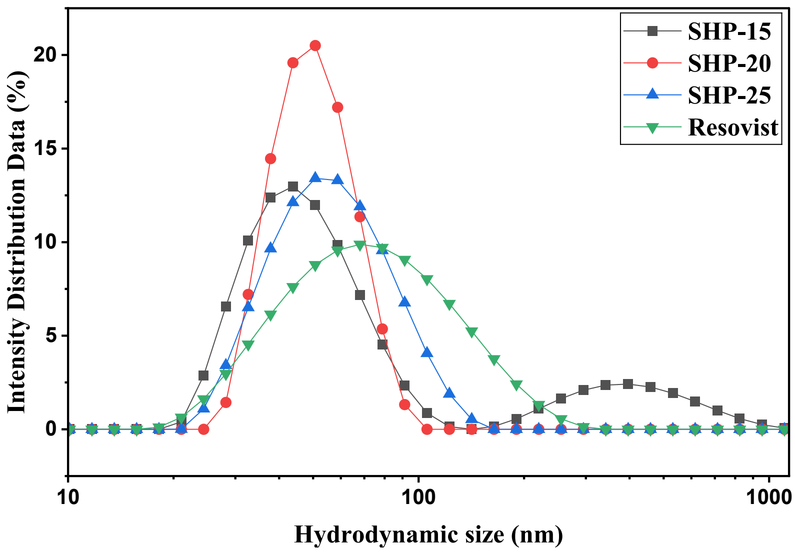

Fig. S4 Hydrodynamic size distribution of SHP-15, SHP-20, SHP-25, and Resovist estimated by DLS.

Table S1 Comparison between the main core size (d_{c_main}) and main hydrodynamic size (d_{h_main}) obtained from VSM and DLS measurements for SHP-15, SHP-20, SHP-25, and Resovist.

Note: Resovist exhibits two peaks in its core size distribution, corresponding to d_{c_main} ≈ 8 nm and 23 nm.

Sample	d_{c_main} [nm]	d_{h_main} [nm]
SHP-15	15	43
SHP-20	20	50
SHP-25	20	55
Resovist	8 and 23	70

TEM images of MS3 and SHP-20

Transmission electron microscopy (TEM) observation was performed using a JEOL 2100F system (JEOL Ltd., Japan) operated at an accelerating voltage of 200 kV.

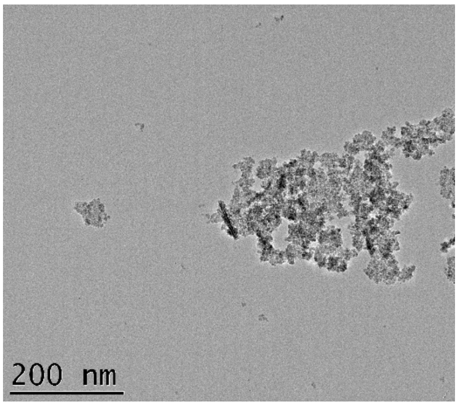

Fig. S5 TEM image of Resovist.

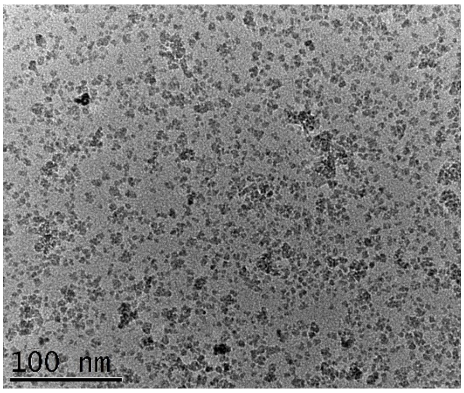

Fig. S6 TEM image of MS3.

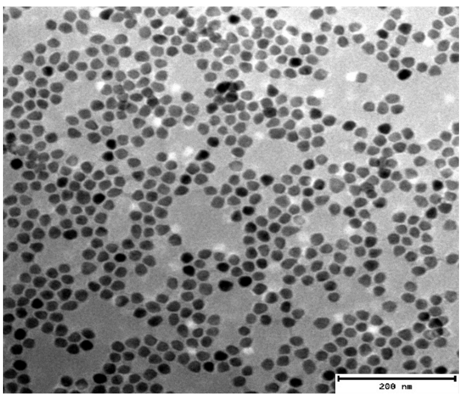

Fig. S7 TEM image of SHP20.

AC *M-H* curves of SHP-15, SHP-25, and Resovist

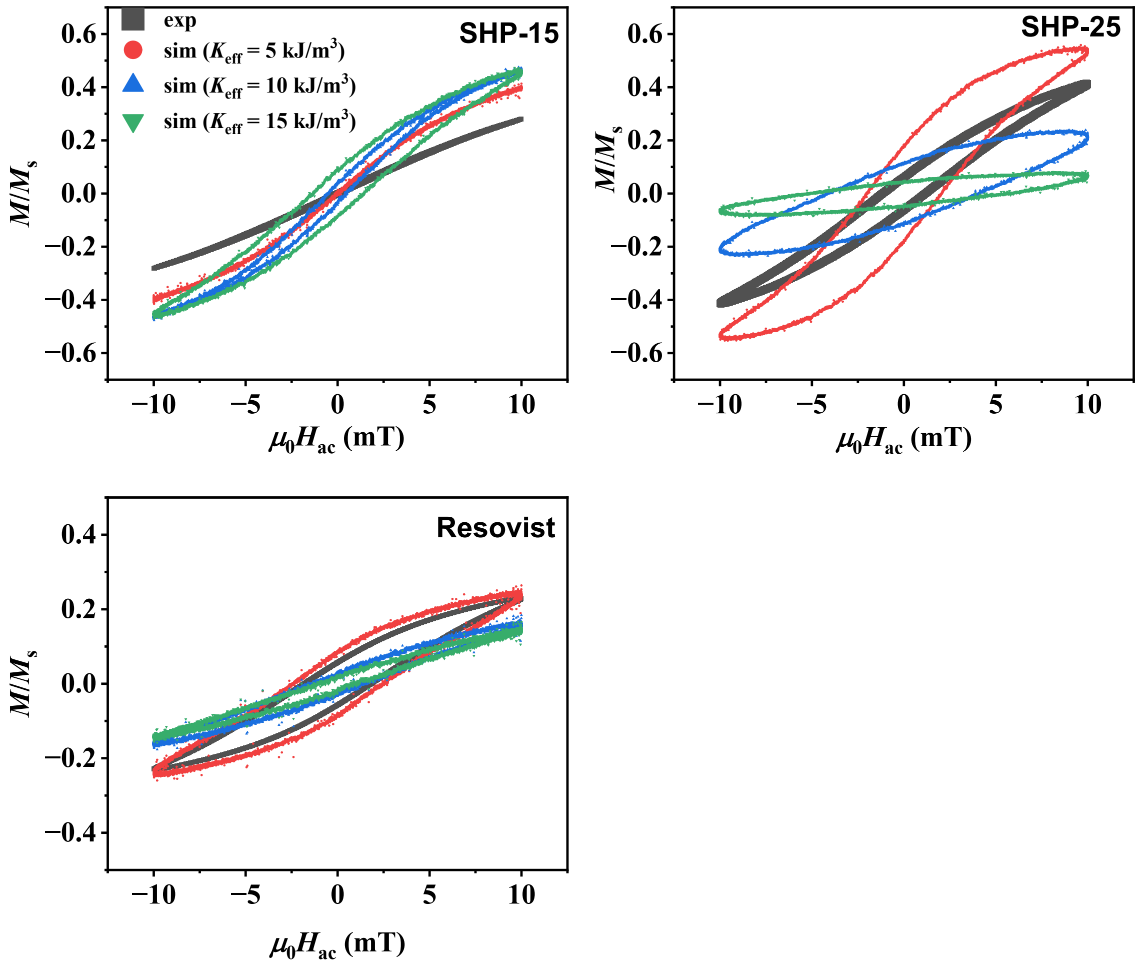

Fig. S8 Comparision between the experimental and simulation results of AC *M-H* for SHP-15, SHP-25, and Resovist under an excitation field amplitude of $\mu_0 H_{ac} = 10\ mT$ and a frequency of 20 kHz. In the simulations, the effective anisotropy constant $K_{eff}(d_c^j)$ is set to a constant value and changed from 5 – 15 kJ/m³.

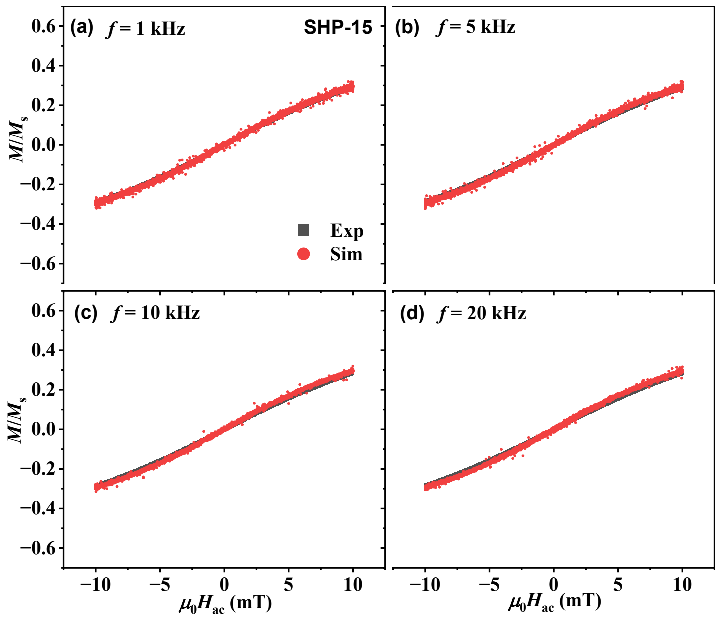

Fig. S9 Comparision between the experimental and simulation results of AC *M-H* for SHP-15 sample under an excitation field amplitude of $\mu_0 H_{ac} = 10\ mT$ and a frequency of (a) 1 kHz, (b) 5 kHz, (c) 10 kHz, and (d) 20 kHz, respectively. In the simulations, the estimated K_{eff} distribution, which is obtained from eqn (12), is used.

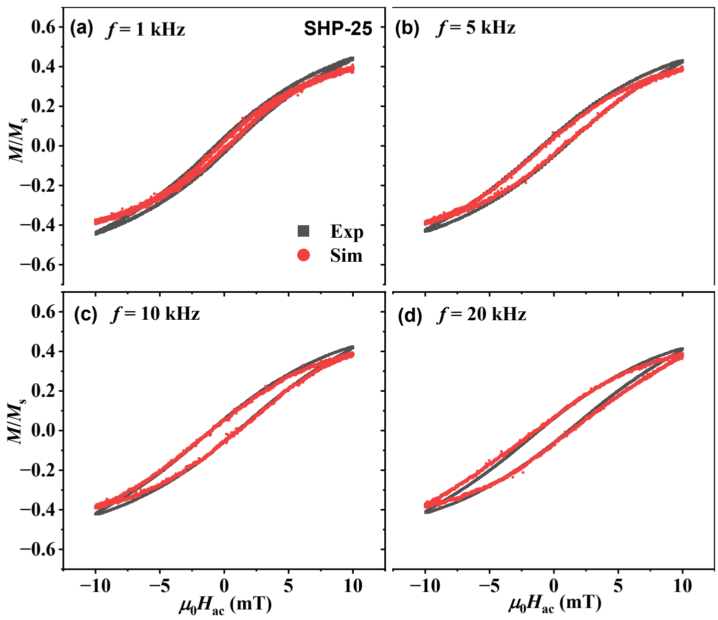

Fig. S10 Comparision between the experimental and simulation results of AC *M-H* for SHP-25 sample under an excitation field amplitude of $\mu_0 H_{ac} = 10\ mT$ and a frequency of (a) 1 kHz, (b) 5 kHz, (c) 10 kHz, and (d) 20 kHz, respectively. In the simulations, the estimated K_{eff} distribution, which is obtained from eqn (12), is used.

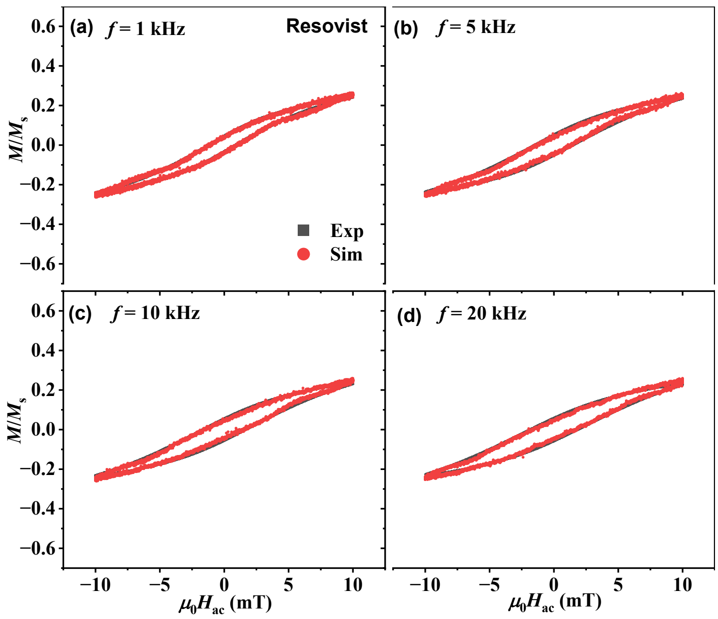

Fig. S11 Comparision between the experimental and simulation results of AC *M-H* for Resovist sample under an excitation field amplitude of $\mu_0 H_{ac} = 10\ mT$ and a frequency of (a) 1 kHz, (b) 5 kHz, (c) 10 kHz, and (d) 20 kHz, respectively. In the simulations, the estimated K_{eff} distribution, which is obtained from eqn (12), is used.

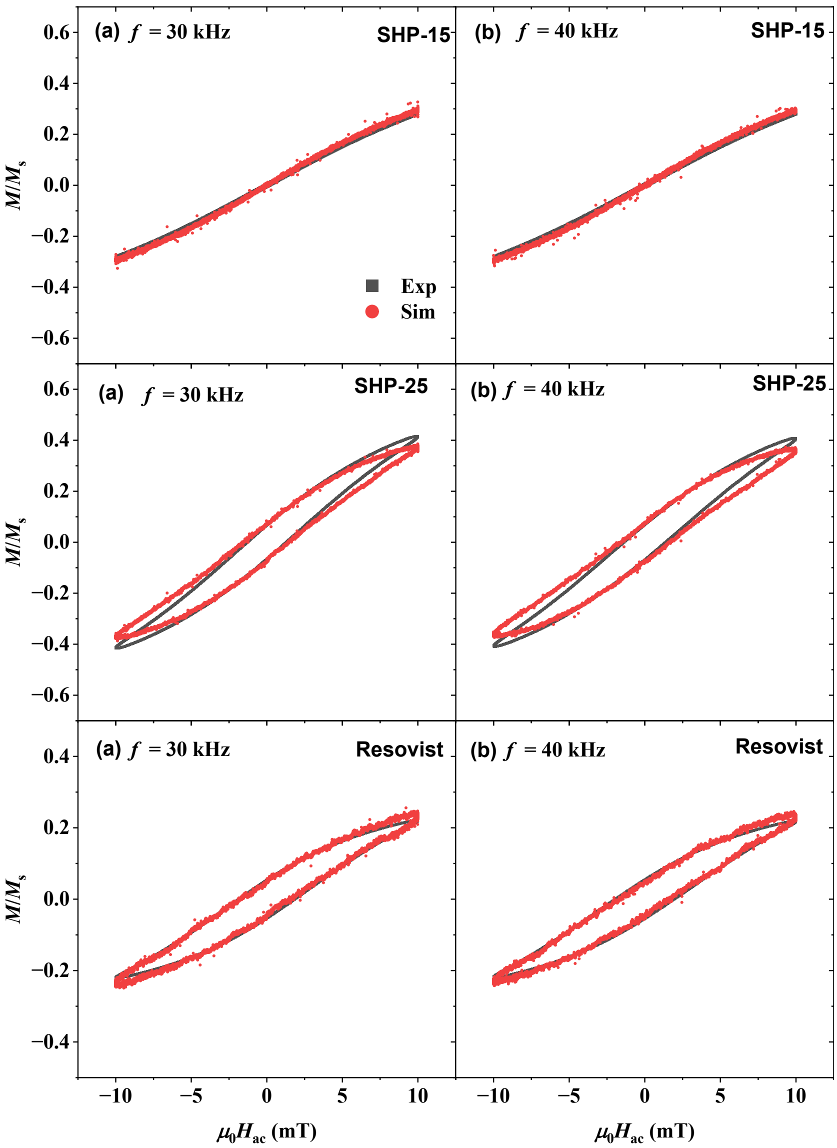

Fig. S12 Comparision between the experimental and simulation results of AC M-H for SHP-15, SHP-25, and Resovist samples under an excitation field amplitude of $\mu_0 H_{ac} = 10\ mT$ and a frequency of (a) 30 kHz and (b) 40 kHz, respectively. In the simulations, the estimated K_{eff} distribution, which is obtained from eqn (12), is used.

K_{eff} **around** d_{c_main}

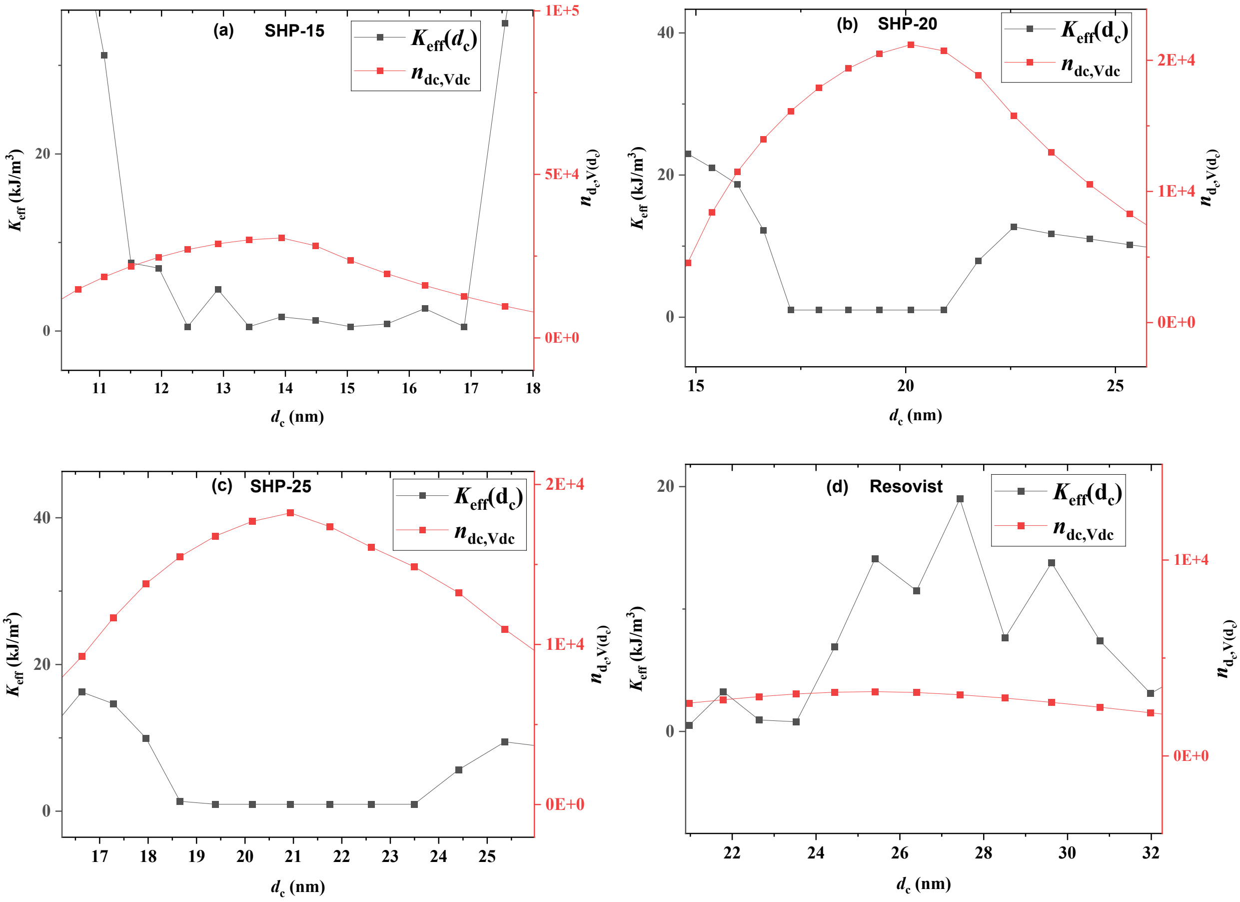

Fig. S13 A partial enlarged view of Figure 4(a-d), when d_c around d_{c_main}.

Comparison between the experimental and simulation results of hysteresis loop area

$$A = -\frac{\mu_0}{M_s}\int_{-H_{ac}}^{H_{ac}} M\,dH \qquad (S1)$$

Table S2 Comparison between the experimental and simulation results of hysteresis loop area *A*, for SHP-15, SHP-20, SHP-25, and Resovist, under an excitation field amplitude of $\mu_0 H_{ac} = 10\ mT$ and a frequency f of 30 kHz and 40 kHz. Hysteresis loop area *A* is calculated by eqn (S1). In the simulations, the estimated K_{eff} distribution, which is obtained from eqn (12), is used.

Sample	A_{exp} (f = 30 kHz) [arb unit]	A_{sim} (f = 30 kHz) [arb unit]	A_{exp} (f = 40 kHz) [arb unit]	A_{sim} (f = 40 kHz) [arb unit]
SHP-15	0.17564	0.1525	0.18965	0.22574
SHP-20	2.65218	2.64797	2.71673	2.88557
SHP-25	1.72174	2.07473	1.80636	2.27716
Resovist	1.30608	1.27624	1.35836	1.36283

References

1. K. Enpuku, T. Sasayama, T. Yoshida, *J. Appl. Phys*. 2016, **119** (18), 184902.
2. J. V. Rijssel, B. W.M. Kuipers, B. H. Erné, *J. Magn. Magn. Mater*, 2014, **353**, 110-115,