Supporting Information

Dynamics of magnetic Janus colloids studied by ultra small-angle x-ray photon correlation spectroscopy

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Magnetic field setup

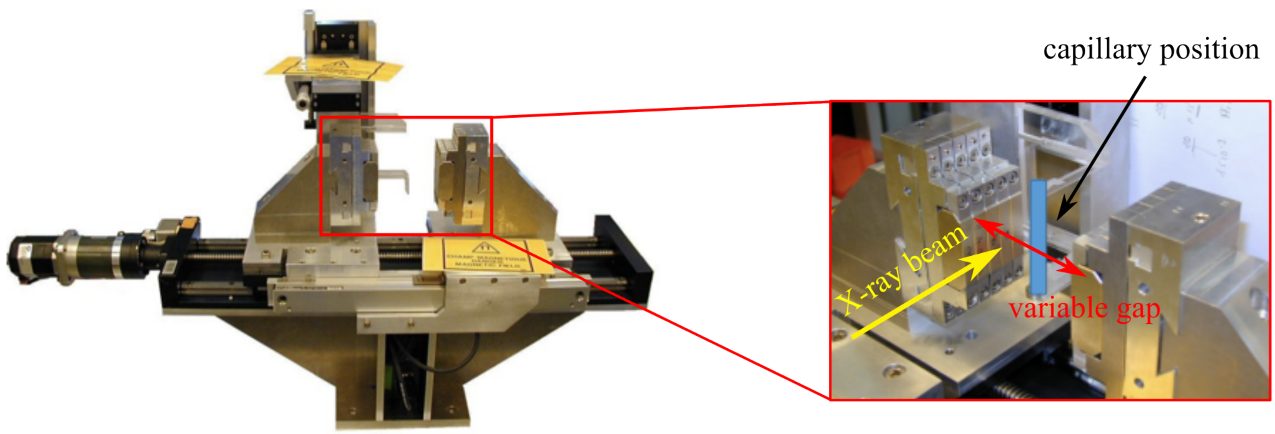

Fig. S1 Permanent magnet setup used for the measurements (left) and the zoom shows the sample position within the Halbach arrangement of magnets (right).

2D USAXS pattern from a dilute sample at zero-field

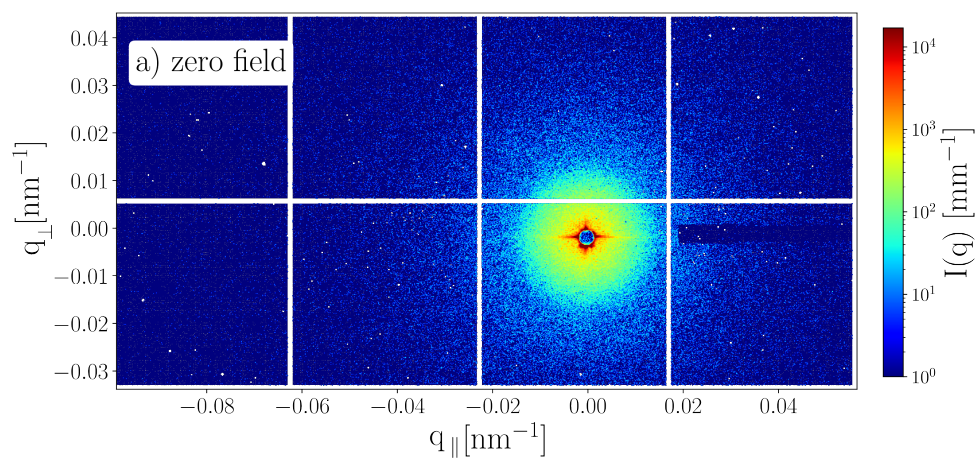

Fig. S2 Measured 2D USAXS pattern from a Janus suspension ($\phi = 7 \times 10^{-4}$) under zero-field i.e. no magnetic field setup installed. The streaks at the lowest q are from the slits and not related to any orientation within the sample.

The field alignment of the dilute sample $\phi = 10^{-4}$

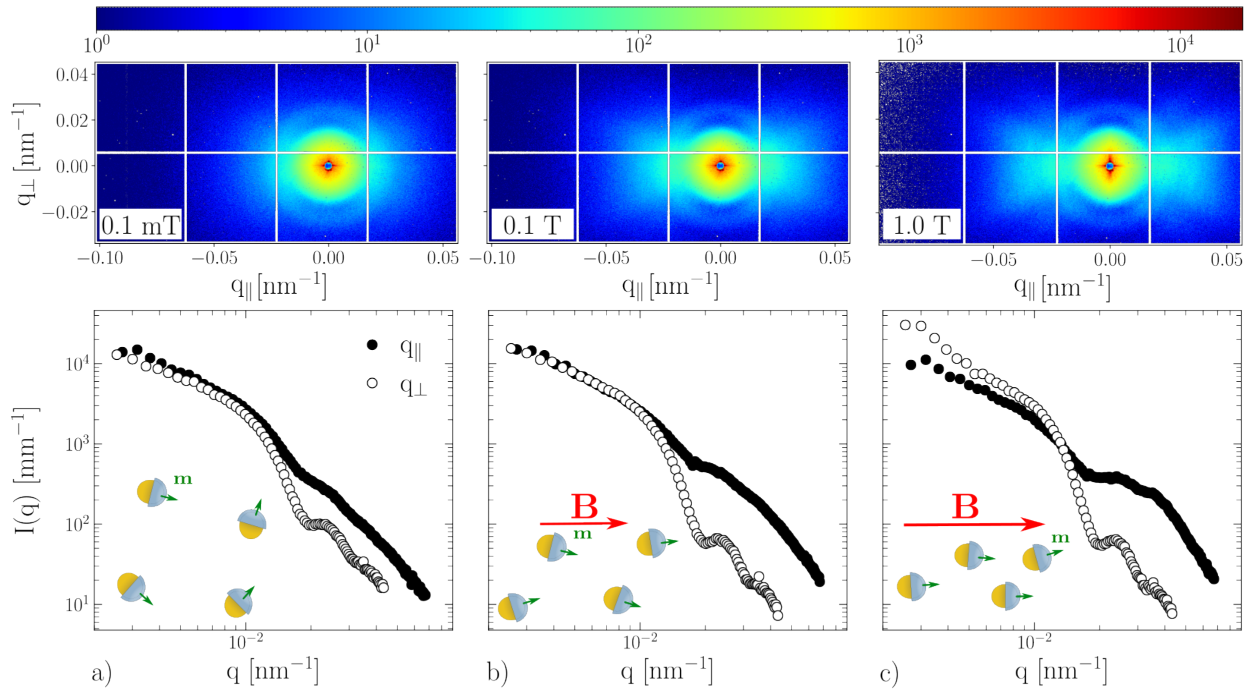

Fig. S3 Measured USAXS intensity from spherical silica-nickel Janus colloids ($\phi = 10^{-4}$) of size about 480 nm when the magnetic field of different flux densities applied perpendicular to the X-ray beam a) 0.1 mT, b) 0.1 T and c) 1 T. For the anisotropic scattering, cuts of the 2D patterns were taken in the direction parallel and perpendicular to the applied field, and averaged over azimuthal sectors of ± 5°. The 1D scattering profiles $I(q)$ are plotted below the corresponding 2D images.

Alignment behavior of a Janus particle suspension for different directions of the applied magnetic field

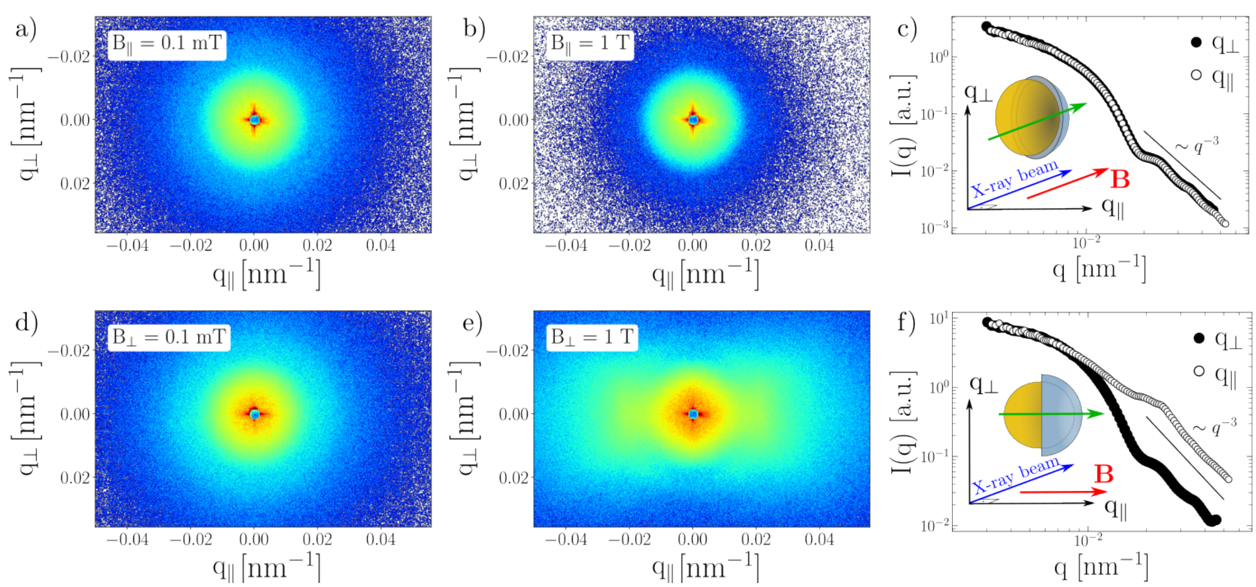

Fig. S4 Observed field direction dependence of particle orientation: a)-c) When the magnetic field is applied along the X-ray beam direction. There is no signature of alignment of particles in the 2D USAXS pattern as also demonstrated by the 1D profiles. d)-f) The magnetic field was perpendicular to both the X-ray beam and the sample capillary. Notice the significant orientation at higher q values.

Simulated USAXS profiles for Janus colloids exposed to magnetic field of 0.1 mT

The 1D USAXS intensity profiles for different concentrations are depicted in figure S5 a) $\phi = 10^{-4}$, b) $\phi = 7 \times 10^{-4}$ and c) $\phi < 10^{-4}$ for the lowest applied magnetic field $B = 0.1\,\mathrm{mT}$. As can be seen the higher ϕ samples show anisotropic scattering that especially becomes notable at higher q-values, whereas for the lowest ϕ the scattering is isotropic, see figure S5c). This is the result of the high sensitivity of the JPs towards magnetic interactions. Consequently, while placing the capillary in the sample holder the JPs feel the magnetic field and start to orient in the direction of magnetic field. Even though special care was taken to prevent pre-exposure to the field. The scattering profiles were attempted to be described in terms of an isotropic form factor model for polydisperse spheres with an hemispherical cap[1]. The model yielded a mean radius of particles $R_s = 180\,\mathrm{nm}$ with a Gaussian polydisperisty of 20% and a thickness of the nickel layer of $d_{\mathrm{Ni}} = 38\,\mathrm{nm}$. The model scattering length densities of SiO_2 and Ni were $1.7 \times 10^{-3}\,\mathrm{nm}^{-2}$ and $6.2 \times 10^{-3}\,\mathrm{nm}^{-2}$, respectively with approximately 10% lower density than bulk amorphous silica. The scattering profiles at the selected concentrations are similar which is an indication for non-interacting particles ($S(q) \approx 1$) and thus mainly magnetic interactions influence the system.

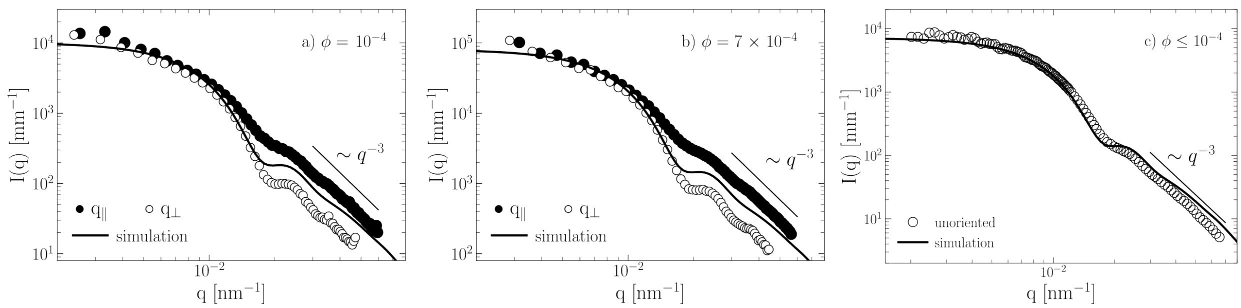

Fig. S5 Simulated USAXS intensity profile for non-interacting particles ($S(q) \approx 1$) for the a) diluted sample ($\phi = 10^{-4}$), b) the concentrated sample ($\phi = 7 \times 10^{-4}$) and c) an unoriented sample $\phi < 10^{-4}$. The solid lines display the modelling in terms of the isotropic scattering form factor for of polydisperse spheres with a hemispherical cap ($R_s = 180\,\mathrm{nm}$, PD $\simeq 0.2$, $d_{\mathrm{Ni}} = 38\,\mathrm{nm}$).

Dynamics of the dilute sample $\phi = 10^{-4}$

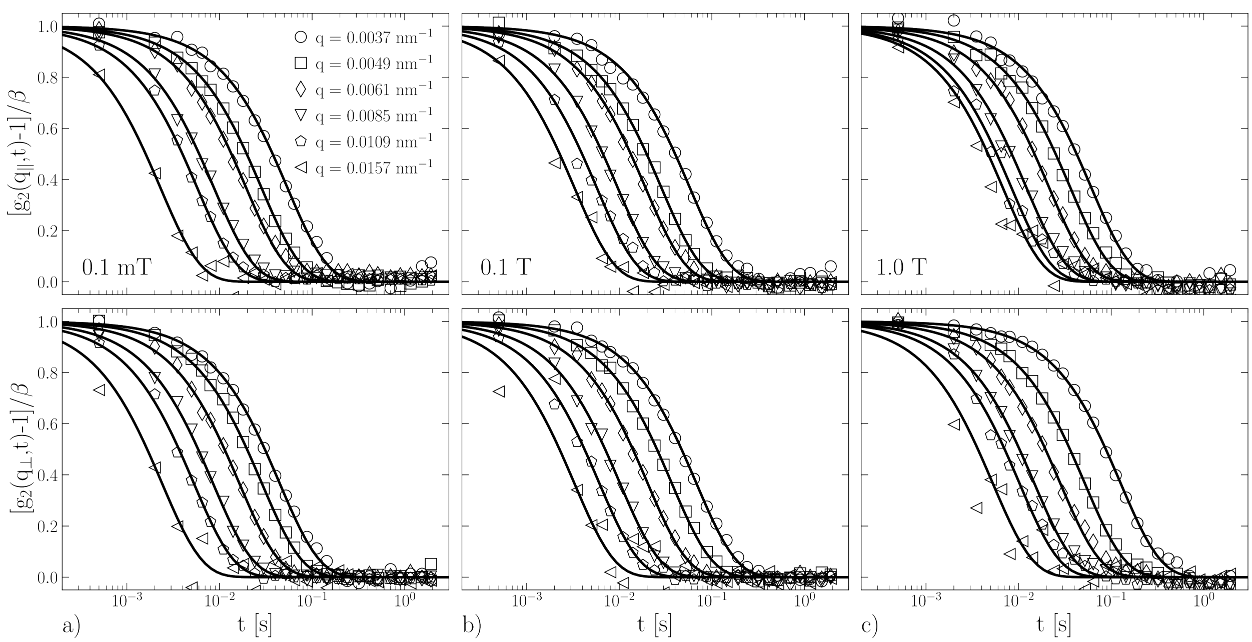

Fig. S6 Normalized intensity-intensity autocorrelation functions $g_2(q,t)$ for a range of q-values indicated for magnetic fields a) 0.1 mT, b) 0.1 T and c) 1.0 T. Solid lines are non-linear-least square fits according to equations given in the main text.

Fit parameters for bimodal decay of $g_2(q,t)$ at $1.0\,\mathrm{T}$ for the concentrated sample, $\phi = 7 \times 10^{-4}$.

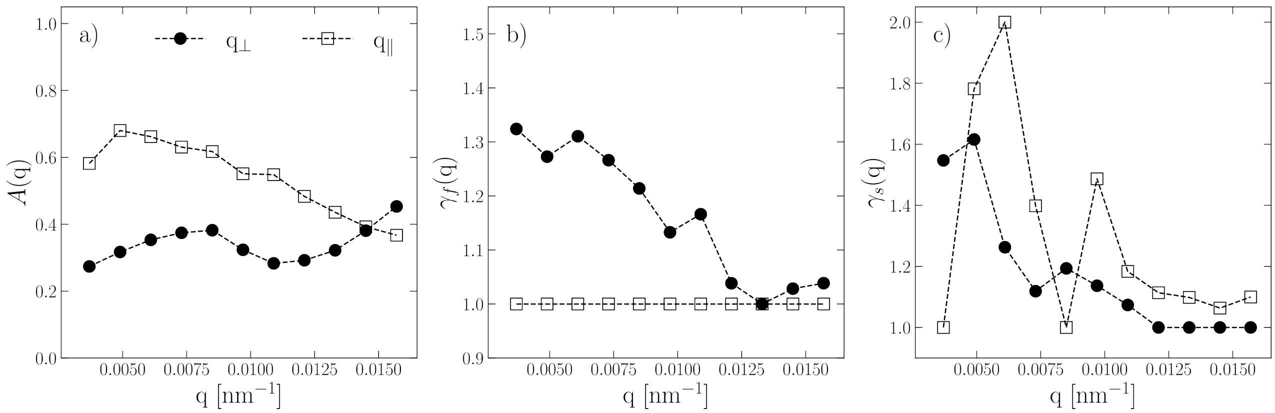

Fig. S7 Obtained fit parameters a) A and the Kohlrausch exponents b) γ_f and c) γ_s, respectively, for the two-step relaxation observed with the concentrated sample in $1.0\,\mathrm{T}$ magnetic field.

Notes and references

1 E. F. Semeraro, R. Dattani and T. Narayanan, J. Chem. Phys., 2018, **148**, 014904.