Supplementary Material (ESI) for Soft Matter

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Electronic Supporting Information (ESI)

Diffusive Dynamics of Nanoparticles in Aqueous Dispersions

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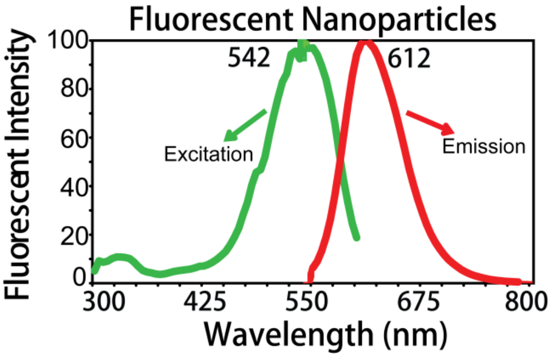


FIG.S.1: Fluorescent spectrum for nanoparticle dispersions at weight fraction of 1%. The maximum excitation and emission wavelengths are 542 nm and 612 nm respectively. (Original data measured by Specialty Diagnostics Group, Thermo Scientific.)

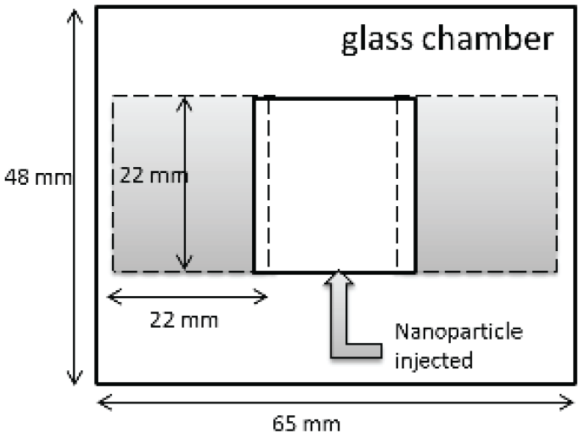


FIG.S.2: Schematic diagram of glass chamber used in DDM measurements, which consists of three 22×22 mm (thickness 0.19-0.23 mm) Fisherbrand microscopy cover glasses and one 48×65 mm (thickness 0.13-0.17 mm) Gold Seal® cover glass.

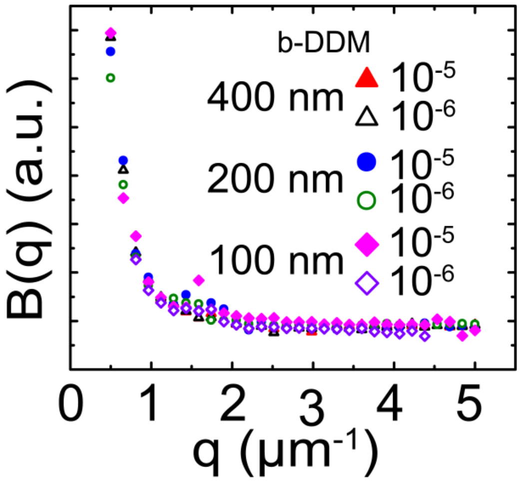


FIG.S.3: The background term *B(q)* as a function of wave vector q obtained by fitting the brightfield-DDM data to Equation (4). The values of *B(q)* agree for samples with low concentration of nanoparticles i.e., $\phi = 10^{-5}$and 10^{-6}, indicating that *B(q)* is independent of the concentration and diameter of the nanoparticles and only dependent on the optical elements.

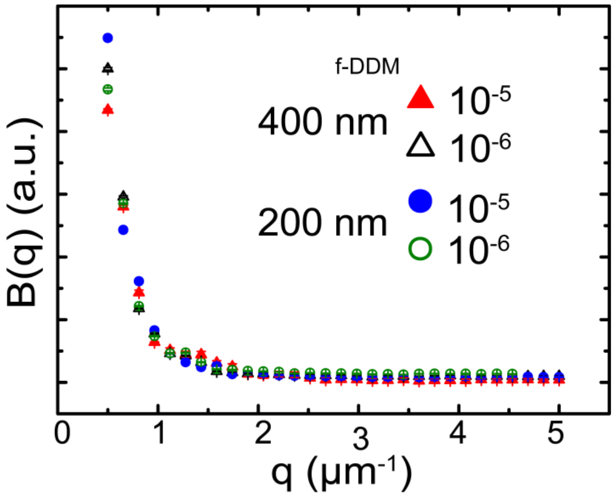


FIG.S.4: The background term *B(q)* as a function of wave vector q obtained by fitting the fluorescence-DDM data to Equation (4). As with the b-DDM, the values of *B(q)* agree for samples with low concentration of nanoparticles i.e., $\phi = 10^{-5}$ and 10^{-6}, indicating that *B(q)* is independent of the concentration and diameter of the nanoparticles and only dependent on the optical elements. We note that the f-DDM for the dispersions of 100 nm particles at ϕ of $10^{-5}$and 10^{-6} did not yield signals with sufficient signal to noise (as shown in Fig S.8). Further, we note that the values of *B(q)* obtained in b-DDM (Fig S.3) and f-DDM are different and not related to each other.

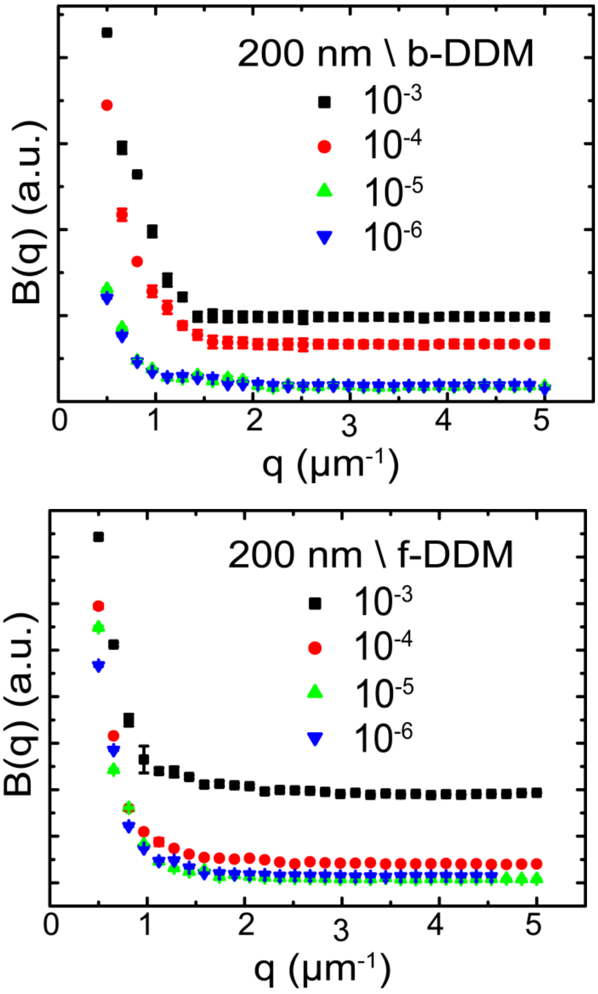
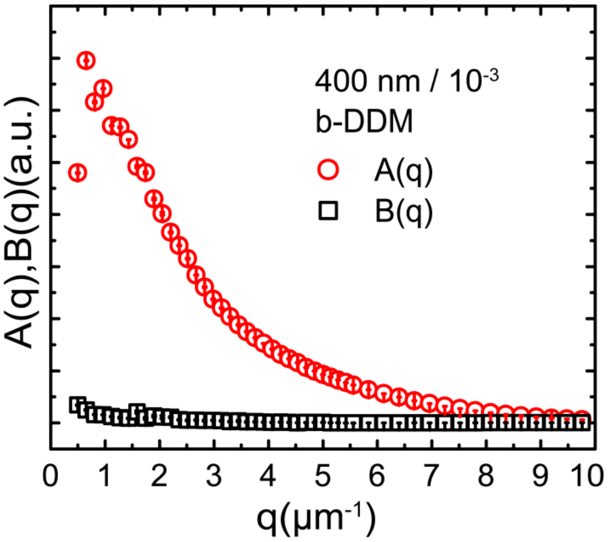


FIG.S.5: The background term *B(q)* as a function of wave vector q obtained by fitting the b-DDM (Top) and f-DDM (Bottom) data to Equation (4) for the aqueous dispersions of 200 nm nanoparticles at concentrations (ϕ) ranging from 10^{-6} to 10^{-3}. The values of *B(q)* for the higher concentration samples (i.e., $\phi = 10^{-4}$ and 10^{-3}) exceed those obtained from the low concentration samples shown in Figures S.3 and S.4. We note that the relaxation times obtained and the diffusion coefficients derived using these best fit derived values of *B(q),* or the *B(q)* imposed from lower concentration measurements or arbitrarily setting *B(q)* to 0, are within the errors of the measurement equal and therefore indicating the relative insensitivity of the fitting, at least at high concentrations, to the value of the background intensity.



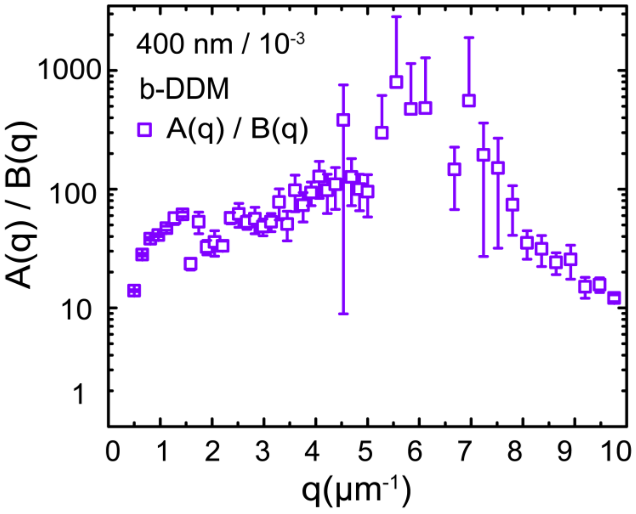
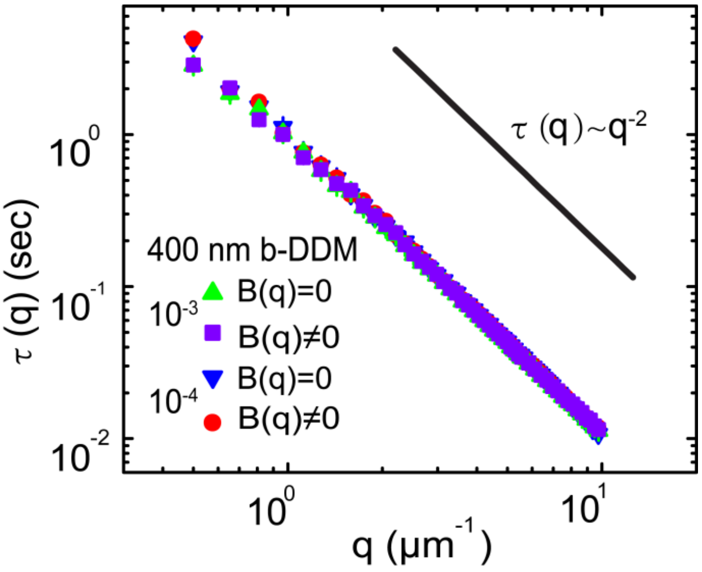


FIG.S.6: Fitting parameters *A(q)* (red circles) and *B(q)* (black squares) (top) and their ratio (bottom) as function of wave vector q obtained by fitting the b-DDM data to Equation (4). Data shown here correspond to fitted parameters obtained for an aqueous dispersion of 400 nm nanoparticles at a volume fraction of 10^{-3}. The value of the signal prefactor *A(q)* is much larger than the value of background term *B(q)* at all values of q for this sample.



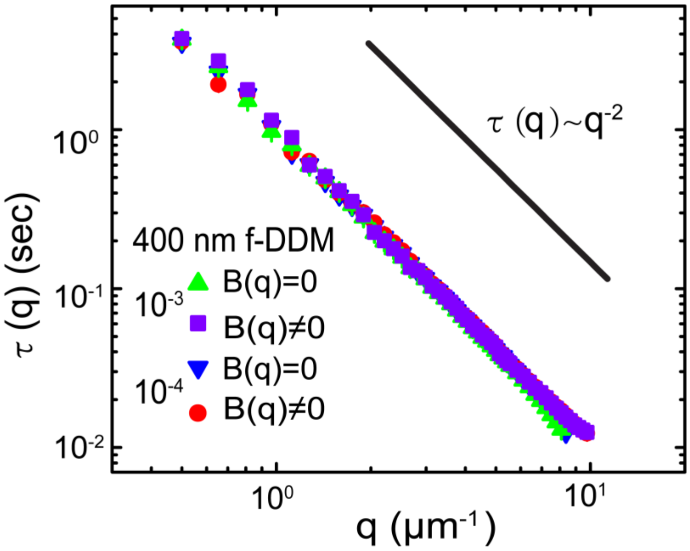


FIG.S.7: Relaxation time τ(q) as a function of wave vector q (μm^{-1}) obtained by fitting the b-DDM (Top) and f-DDM (Bottom) structure functions to Equation (4) for an aqueous dispersion of 400 nm nanoparticles at volume fractions (ϕ) of $10^{-3}$and 10^{-4}. The relaxation times obtained using a best fit value of *B(q)* (which exceeds that obtained from the low concentration samples that were only a function of the optical elements) are within the errors of the measurement identical to those obtained by arbitrarily setting the value of *B(q)* to 0. For the case of the b-DDM (Top), the extracted diffusion coefficients for $\phi=10^{-3}$ using *B(q)*=0 and best fit values of *B(q)* are 0.97±0.08 and 0.96±0.06 μm^2⁄s respectively. Similarly, for the $\phi=10^{-3}$ using *B(q)*=0 and best fit values of *B(q)* the diffusion coefficients are 0.96±0.06, and 0.94±0.05 μm^2⁄s respectively.

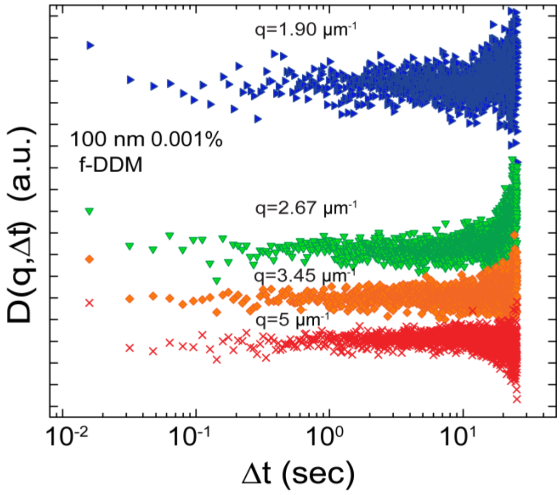


FIG.S.8: Structure function *D(q, Δt)* as a function of delay time Δt at wavevectors q of 1.9 μm^{-1}(blue), 2.67 μm^{-1}(green), 3.45 μm^{-1}(orange), and 5 μm^{-1}(red) acquired using f-DDM for an aqueous dispersion of 100 nm diameter nanoparticles at ϕ of 10^{-5}. *D(q, Δt)* does not exhibit an exponential increase, which indicates that f-DDM cannot be used to measure diffusive dynamics of this sample due to the extremely low signal compared with the background.