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Probing of Pair Interaction of Magnetic Microparticles with Optical Tweezers¶

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Magnetic interaction of paramagnetic Brownian submicron-sized particles is studied by optical tweezers technique. Correlation analysis allows one to extract magnetic interaction of two particles 0.4 μm in size, which are optically trapped at the distance of 3 μm one from each other and placed in a static magnetic field of 30 Oe, from the background of their Brownian motion. The magnetic interaction force is estimated to be of approximately 100 fN. Two configurations of the mutual orientation of the magnetic field vector and the line connecting two centers of optical traps are used in the experiment. For field vector orientation parallel/perpendicular to this line, the magnetic interaction is detected by the cross-correlation function increase/decrease in comparison with the absence of magnetic field on the time scales of 1 ms.

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Magnetic liquids, which are suspensions of magnetic microparticles, have been actively studied in recent decades. These suspensions have numerous practical applications, such as the production of magnetic memory devices [1] and cancer treatment by the hyperthermia method [2]. Single magnetic microparticles are used for sensitive monitoring of the properties of media [3, 4] and as an instrument allowing one to manipulate single biological cells and macromolecules. As an example, the DNA torsion modulus was measured using magnetic microparticles [5]. The collective behavior of magnetic microparticles in an external alternating magnetic field was studied in numerous works [6, 7]; however, the problem of pair interaction between magnetic microparticles in suspension was discussed only in a few papers [4, 8]. Since for these systems magnetic interaction forces on the micron spatial scales are on the order of 100 fN, it is reasonable to use optical tweezers for measuring weak magnetic forces [9, 10]. Optical tweezers are based on the formation of a potential well for transparent dielectric microobjects located near the focus of the tightly focused laser beam that allows trapping of microobjects and manipulation of them. Optical tweezers have a lot of applications for studying force interactions between biological cells [11], measuring elastic characteristics of biological cell membranes and individual macromolecules [12], and studying luminescent [13] and nonlinear-optical [14] properties of single microparticles. This method was also used for studying the behavior of magnetic microparticles in a rotating magnetic field [15] and measuring the magnetic moments of trapped particles [8]. However, detection of weak magnetic forces between microparticles remains a complicated problem because of Brownian motion. In this paper, a cross-correlation function analysis is proposed for detection of the magnetic interaction force between two trapped particles. Previously, this method was used for studying the hydrodynamic interaction between microparticles [16].

Two optically trapped microparticles interact by means of the liquid medium in which they are dipped. Their equations of motion are as follows:

$$\frac{d\mathbf{R}_n}{dt} = \sum_{m=1}^{2} \mathbf{\Pi}_{nm}(\mathbf{R}_n - \mathbf{R}_m)[\mathbf{k}\mathbf{r}_m + \mathbf{F}_m(t)], \qquad (1)$$

where n, $m = 1, 2$ are the particle numbers; $\mathbf{R}_n$ is the particle radius vector; $\mathbf{F}_n(t)$ is the stochastic Brownian force acting on the nth particle; $\mathbf{k}$ is the effective stiffness tensor determining trapping force $\mathbf{k}\mathbf{r}_n$, which appears when the nth particle is displaced by the distance $\mathbf{r}_n = \mathbf{R}_n - \mathbf{R}_{0n}$ from the optical trap center $\mathbf{R}_{0n}$; and $\mathbf{\Pi}_{nm}$ is the Oseen tensor. Its elements can be written as follows:

$$\mathbf{\Pi}_{nn}(\mathbf{R}) = \frac{\mathbf{I}}{\gamma}, \quad \mathbf{\Pi}_{nm}(\mathbf{R}) = \frac{\mathbf{I} + \hat{\mathbf{R}}\hat{\mathbf{R}}}{8\pi\eta R}, \qquad (2)$$

where $\gamma = 3\pi\eta d$ is the friction coefficient for a spherical particle with the diameter d in a liquid medium with the viscosity η, $\mathbf{I}$ is a 3×3 unit matrix, $\hat{\mathbf{R}}$ is the unit vector parallel to the vector connecting particle position points $\mathbf{R} = \mathbf{R}_2 - \mathbf{R}_1$, and $R = |\mathbf{R}|$ is the distance between the particles. Solution of Eqs. (1) gives

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the expression for the cross-correlation function for vector components of particle positions $\mathbf{r}_n$ [16]:

$$\langle r_{1,i}(0)r_{2,i}(t)\rangle = \frac{k_B T}{2k_i}[e^{-t(1+\varepsilon_i)k_i/\gamma} - e^{-t(1-\varepsilon_i)k_i/\gamma}], \quad (3)$$

where $i = x, y, z$, angle brackets denote time averaging, $k_B T$ is the product of the Boltzmann constant and absolute temperature, and k_i is the effective trap stiffness in the direction of the i axis. If the line connecting the traps centers is parallel to the x axis, then $\varepsilon_x = 3d/4L$, and $\varepsilon_y = \varepsilon_z = 3d/8L$, where L is the distance between the trap centers. The minimum of the cross-correlation function is observed at times of about $\tau_i = \gamma/k_i$. It is equal to $-k_B T\varepsilon_i/ek_i$ and is induced by hydrodynamic interaction between the particles.

Additional interaction between microparticles, for instance magnetic interaction, leads to the displacement of particles from equilibrium positions in optical traps. This displacement has a magnitude on the order of $F_{magn}/k_i \sim 10^{-6}$ cm. The equipartition theorem for an object in a harmonic potential with stiffness k_i can be written as $k_i\langle r_{n,i}^2\rangle/2 = k_B T/2$ [10], and the root-mean-square displacement of particle Brownian motion is estimated to be $\sqrt{\langle r_{n,i}^2\rangle} \sim 10^{-6}$ cm. Therefore, particle displacement from the optical trap center due to Brownian motion is comparable with displacement under magnetic force action. This complicates direct measurement of magnetic forces between particles. However, magnetic forces might change the shape of the cross-correlation function of Brownian displacement of particles. Indeed, magnetic moments of microparticles lead to either attraction or repulsion of particles. These attractive or repulsive forces change the probability of Brownian displacements of particles from the equilibrium positions in optical traps. In the case of attractive force, the first particle displacement to the right (see Fig. 1a) leads to the increase in the attractive force acting on the second particle. Therefore, the probability of the second particle to be displaced to the left increases, which leads to the decrease in the cross-correlation function. In the case of repulsive forces between the particles, the cross-correlation function increases. The first particle displacement to the right induces the increase in the repulsive force acting on the second particle, which will more probably be shifted to the right (see Fig. 1b).

In the present paper, the interaction between paramagnetic microparticles is studied by correlation function analysis combined with optical tweezers technique. Interaction is detected by magneto-induced changes in the cross-correlation function of Brownian displacements of trapped particles.

Experimental samples were a water suspension of composite paramagnetic particles with a mean size of 0.4 μm made from silica doped with ferric oxide (III) (Sileks, Russia). The experimental setup of double trap optical tweezers with magnetic field application is shown in Fig. 2. Optical traps were formed by tightly focusing the radiation from two infrared CW lasers with the wavelength of 1064 nm into the sample area. The sample suspension was placed between two coverslips separated by 0.15-mm-thick gap. The laser beam power in the sample area was approximately 10 mW per trap. The root-mean-square amplitudes of Brownian motion displacements of particles were equal to approximately 10 nm, which was sufficient for registration of particle displacements by the data acquisition system. The system of lenses made it possible to control the positions of optical traps in the sample area by moving the lenses perpendicular to the beam propagation direction. An external magnetic field was applied to the sample using a system of electromagnets consisting of four independent coils each having a tip pole expansion. A homogeneous magnetic field with different magnetic vector orientations was created by controlling the current through each coil. The field strength was about 30 Oe. For visualization of the experiment, white LED radiation passed through the condenser, illuminated the sample, and then was registered using a CCD camera. A typical picture of two trapped microparticles is shown in the inset of Fig. 3. Position-sensitive diodes (PSDs) were used for particle displacement measurement in real time with precision of about 1 nm. Signals from the PSDs were pro-

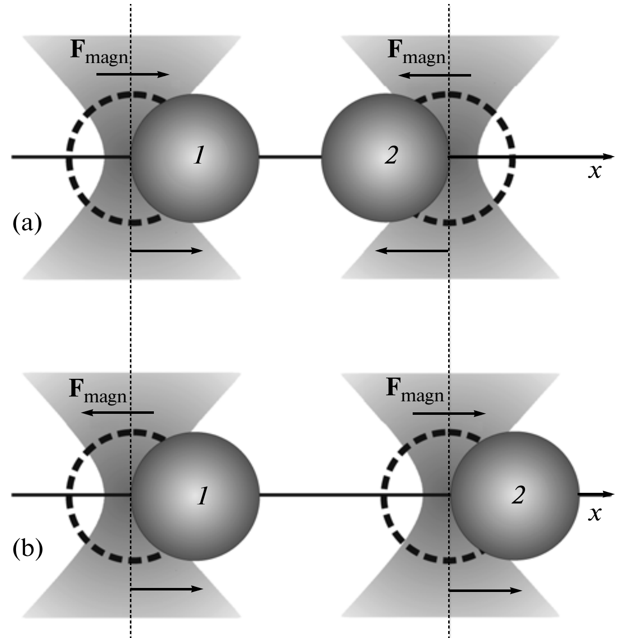

Fig. 1. Schematic of the influence of magnetic particle interaction on cross-correlations in Brownian motion of particles in optical traps; $\mathbf{F}_{magn}$ is magnetic interaction force; forces of (a) attraction and (b) repulsion act between particles.

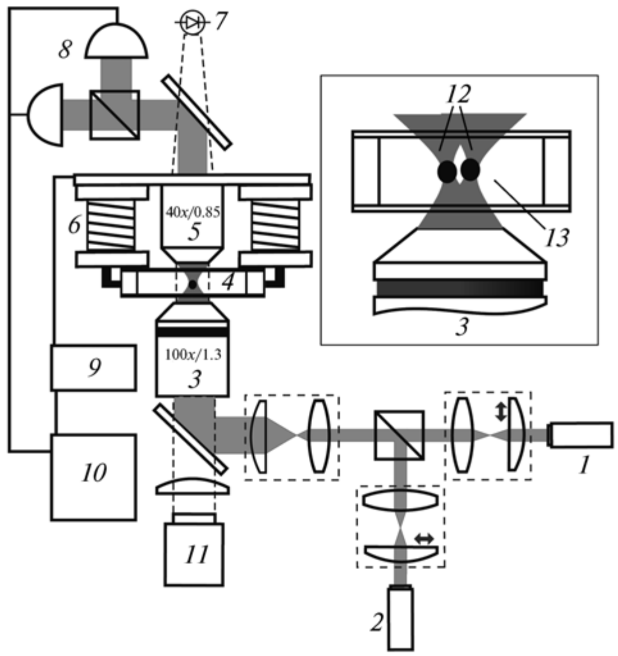

Fig. 2. Schematic of the setup of the double trap optical tweezers: (*1*, *2*) YAG-Nd lasers; (*3*) objective lens; (*4*) chamber with paramagnetic suspension of microparticles; (*5*) condenser lens; (*6*) system of electromagnets for magnetic field application; (*7*) LED, (*8*) position-sensitive photodiodes; (*9*) current boost; (*10*) analog-to-digital and digital-to-analog converters; (*11*) CCD camera; (*13*) two magnetic microparticles trapped in two optical traps *12*.

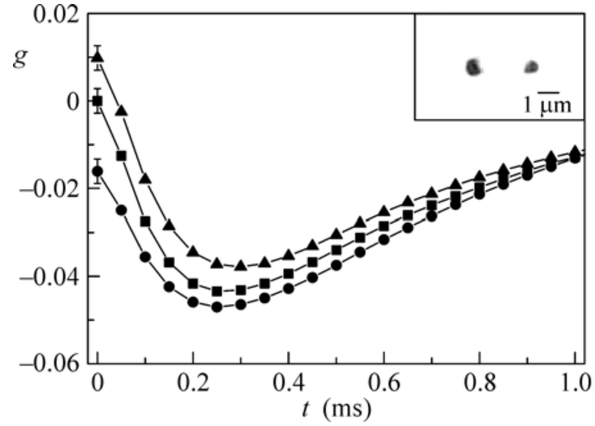

Fig. 3. Time dependences of normalized cross-correlation functions of Brownian displacements for two optically trapped particles: squares are for the absence of magnetic field, circles are for the external magnetic field vector with the magnitude of 30 Oe oriented parallel to the line connecting the two laser traps, and triangles are for the magnetic field vector of the same value and oriented perpendicular to this line. Inset: microimages of two trapped microparticles.

cessed by an analog-to-digital converter with a sampling rate of 20 kHz [10].

In the experiment, two magnetic microparticles were optically trapped at the distance of 10 μm above the lower coverslip of the chamber. The distance L between the particles was fixed at 3 μm. First, the statistics of Brownian displacements of the particles were recorded for 100 s without an external magnetic field, and the cross-correlation functions of the displacements were determined. In this case, the magnetic moments of particles and their magnetic interaction forces were negligible and the cross-correlation functions were similar to those of dielectric beads given by Eq. (3). Then cross-correlation functions of Brownian displacements were measured in parallel and perpendicular configurations of external dc magnetic field application with the magnitude of the field strength vector **H** equal to 30 Oe. The magnetic field vector in the parallel configuration was directed along the line connecting the trap centers, i.e., parallel to the x axis. In the perpendicular configuration, it was perpendicular to the x axis, as shown in Fig. 1.

The cross-correlation functions of the particle displacements along the x axis, $B(t) = \langle x_1(0)x_2(t)\rangle$, were analyzed since the projections of the magnetic interaction forces on the x axis were maximal. To account for the particle size dispersion and the impact of the particle form, the cross-correlation functions were normalized to the dispersion of Brownian displacements (Δx^2):

$$g(t) = \frac{B(t)}{\sqrt{\Delta x_1^2 \Delta x_2^2}} = \frac{\langle x_1(0)x_2(t)\rangle}{\sqrt{\Delta x_1^2 \Delta x_2^2}}. \quad (4)$$

Typical normalized cross-correlation functions are shown in Fig. 3. The cross-correlation curves exhibit a time-delayed anticorrelation with a pronounced minimum at 0.3 ms induced by hydrodynamic interaction between the particles. The depth of the minimum is about 0.04 when the magnetic field is absent. This allows one to give estimates of the stiffness of optical traps and the diameter of particles which appear to be $k_x \approx 2 \times 10^{-2}$ dyn/cm and $d \approx 4 \times 10^{-5}$ cm, respectively. A decrease in the cross-correlation functions is observed when the magnetic field vector orientation is parallel to the x axis, meaning that the anticorrelation becomes more pronounced. When the magnetic field vector is perpendicular to the x axis, an increase in the cross-correlation functions is observed, indicating the appearance of correlation of the particle displacements.

In the presence of an external magnetic field, magnetic moments were induced in the particles. In case of magnetic field vector orientation parallel or perpendicular to the x axis, the magnetic particles were oriented so that their magnetic moments were codirectional with each other and the external magnetic field (Fig. 4). In the dipole approximation, the x axis projections of magnetic forces for the magnetic vector parallel to the x axis can be written as follows [4, 8]:

$$F_{\parallel,1} = -F_{\parallel,2} = \frac{6M^2}{R^4}. \quad (5)$$

In the case of magnetic vector orientation perpendicular to this axis, the magnetic force projections are as follows:

$$F_{\perp,1} = -F_{\perp,2} = -\frac{3M^2}{R^4}, \quad (6)$$

where M is the value of the microparticle magnetic moment. If particle magnetic moments are perpendicular to the line connecting the trap positions, the repulsive forces appear between the particles. In the case of orientation of the magnetic moments parallel to the line connecting the trap positions, the attractive forces take place. For the same magnetic moment values, the attractive force magnitude is double the repulsive force value. With regard to the magnetic interaction forces, the equations of particle motion can be written as follows:

$$\frac{d\mathbf{R}_n}{dt} = \sum_{m=1}^{2} \mathbf{\Pi}_{nm}(\mathbf{R}_n - \mathbf{R}_m) \times [\mathbf{k}\mathbf{r}_m + \mathbf{F}_m(t) + \mathbf{F}_{\text{magn},m}], \quad (7)$$

where $\boldsymbol{F}_{\text{magn},n}$ is the magnetic interaction force acting on the nth particle. Neglecting the changes in the distance between the particles induced by their Brownian motion along the y and z axes, we consider that the distance between the particles can be written as $R = L + x_2 - x_1$, where x_1 and x_2 are the x-axis projections of Brownian displacements of particles. Therefore, the impact of magnetic forces on the cross-correlation functions can be estimated. The Brownian displacements of particles are on the order of tens of nanometers; i.e., they are much less than the distance between the particles that is on the order of several microns. Therefore, magnetic forces (5) and (6) can be expanded in the small parameter $\Delta/L \equiv (x_2 - x_1)/L$ (first two terms of the expansions are retained):

$$\begin{aligned} F_{\parallel,1} &= -F_{\parallel,2} \approx \frac{6M^2}{L^4} - \frac{24M^2}{L^5}\Delta, \\ F_{\perp,1} &= -F_{\perp,2} \approx -\frac{3M^2}{L^4} + \frac{12M^2}{L^5}\Delta. \end{aligned} \quad (8)$$

Each of Eqs. (8) consists of two components. The first component is independent of Brownian displacements and proportional to M^2/L^4. This force component displaces the particles from the equilibrium positions in the optical trap. The second component depends on the Brownian displacements and therefore

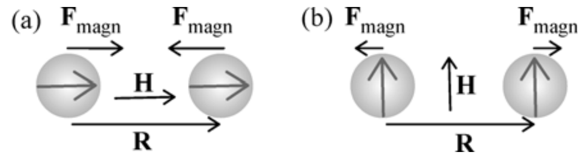

Fig. 4. Schematic diagram of mutual orientation of magnetic moments and forces. The magnetic field vector is (a) parallel and (b) perpendicular to the line connecting the optical trap centers; $\mathbf{F}_{\text{magn}}$ is the magnetic interaction force, $\mathbf{H}$ is the external magnetic field vector, and $\mathbf{R}$ is the vector connecting the positions of particles. Magnetic moments are shown by gray arrows.

influences the form of the cross-correlation function of displacements:

$$B_{\text{magn}}(t) = \frac{k_B T}{2}\left[\frac{e^{-t(1+\varepsilon_x)k_x/\gamma}}{k_x} - \frac{e^{-t(1-\varepsilon_x)k'_x/\gamma}}{k'_x}\right], \quad (9)$$

where $k'_x = k_x - 48M^2/L^5$ if the external magnetic field vector is parallel to the x axis and $k'_x = k_x + 24M^2/L^5$ if it is perpendicular to the x axis. In the presence of a magnetic field, the dispersion of the Brownian displacements is as follows: $\Delta x^2 = k_B T(k_x + k'_x)/2k_x k'_x$. The largest magneto-induced changes in the cross-correlation function are observed for $t = 0$. Consider the expression for the difference $\Delta g(0) = g_{\text{magn}}(0) - g(0)$ revealing the magneto-induced changes in the normalized cross-correlation functions. For parallel and perpendicular magnetic field configurations, it can be written as follows:

$$\begin{aligned} \Delta g_{\parallel}(0) &= -\frac{24M^2}{L^5 k_x - 24M^2}, \\ \Delta g_{\perp}(0) &= \frac{12M^2}{L^5 k_x + 12M^2}. \end{aligned} \quad (10)$$

According to expressions (10), the normalized cross-correlation function change is negative for the attractive forces and positive for the repulsive forces, and $|\Delta g_{\parallel}(0)| > |\Delta g_{\perp}(0)|$, which is in agreement with the experimental results. Cross-correlation function changes can be estimated as $|\Delta g(0)| \simeq 0.01$ and the magnetic moment value is approximately $M \approx 5 \times 10^{-11}$ erg/G (see Fig. 3). Magnetic interaction forces are estimated using Eqs. (5) and (6). The values of the forces are approximately equal to 10^{-8} dyn, i.e., on the order of 100 fN.

In conclusion, the presence of magnetic interaction considerably changes the statistical properties of the Brownian motion of particles. Namely, the cross-correlation function of Brownian displacements of two optically trapped paramagnetic particles changes considerably in the presence of an external magnetic field. The changes in the cross-correlation functions are dependent on the values and directions of magnetic interaction forces. Therefore, the suggested method allows detecting weak magnetic forces on the order of femtonewtons, whereas the direct measurement of these forces is complicated because of the presence of stochastic Brownian motion. Such particle interaction forces considerably affect various processes in magnetic suspensions, such as magnetic particle aggregation, magnetic fluid flow, and magnetic field induced structure formation.

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