Optomagnetically Controlled Microparticles Manufactured with Glancing Angle Deposition

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Optical trapping and magnetic trapping are common micromanipulation techniques for controlling colloids including micro- and nanoparticles. Combining these two manipulation strategies allows a larger range of applied forces and decoupled control of rotation and translation; each of which are beneficial properties for many applications including force spectroscopy and advanced manufacturing. However, optical trapping and magnetic trapping have conflicting material requirements inhibiting the combination of these methodologies. In this paper, anisotropic microscaled particles capable of being simultaneously controlled by optical and magnetic trapping are synthesized using a glancing angle deposition (GLAD) technique. The anisotropic alignment of dielectric and ferromagnetic materials limits the optical scattering from the metallic components which typically prevents stable optical trapping in three dimensions. Compared to the current state of the art, the benefits of this approach are twofold. First, the composite structure allows larger volumes of ferromagnetic material so that larger magnetic moments may be applied without inhibiting the stability of optical trapping. Second, the robustness of the synthesis process is greatly improved. The dual optical and magnetic functionality of the synthesized colloids is demonstrated by simultaneously optically translating and magnetically rotating a magnetic GLAD particle using a custom designed optomagnetic trapping system.

1. Introduction

Design and development of novel particles with anisotropic properties is a burgeoning field of research. Anisotropic particles provide several advantages including enhanced control over position,[1] enhanced tracking capabilities,[2] localized chemical functionality,[3] and functional properties that may be leveraged for sensors[4] or drug delivery.[5] Anisotropic properties may also enhance the manipulability of microscaled particles within optical trapping (OT) or magnetic trapping (MT) systems. Both OT and MT have been extensively demonstrated individually as invaluable micromanipulation techniques. Optical trapping involves the application of an electric field generated by a highly focused laser to impart forces upon micrometer and nanometer scaled particles. Magnetic trapping is a similar procedure that generates forces through the application of an external magnetic field generated by a permanent or electromagnet. Both of these techniques are desirable for many manufacturing, sensor, and experimental procedures because the forces may be applied using externally generated fields that do not require direct access to the sample. The limitations preventing simultaneous application of optical and magnetic trapping (OMT) within a single probe particle is inherently a material compatibility issue. Optical trapping requires dielectrics, which have minimal optical absorption, in order to maximize the OT applied force, prevent undesirable optical scattering forces, and limit thermal vibrations. Alternatively, MT requires the presence of ferromagnetic materials which are not optically transparent and therefore are generally not applicable for OT. In order to combine the benefits of each micromanipulation technique with a single probe particle, the dielectric and ferromagnetic materials must be aligned such that the optical scattering is minimized while still including ferromagnetic material. This paper describes the design and synthesis of particles using a glancing angle deposition (GLAD) process, which results in so-called patchy particles. Due to the material anisotropy, magnetic GLAD particles are compatible with both optical and magnetic-based manipulations.

Previous studies have implemented OT for advanced micro-assembly techniques that could not be achieved using directed self-assembly[6] and for driving micro components within more complex systems.[7] Similarly, anisotropic particles were designed to enhance the magnetic properties of microscaled particles in order to achieve greater magnetic responses and develop magnetic microtools.[8] Particles with improved physical characteristics amenable to OT and MT individually or simultaneously would enable advancements in a wide range of fields including biotechnology and advanced manufacturing. For example, the range of forces and torques applied to a single particle by OT and MT may be varied by over an order of magnitude by simply alternating between optical and magnetic based control.[9] Also, the use of OT for translational manipulation along with MT for rotational manipulation provides holonomic control of a particle, decoupling the application of

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DOI: 10.1002/ppsc.201500033
linear forces and rotational torques. The coupling of force and torque within OT or MT has been a limitation in the field of force spectroscopy due to the nonlinear relationship between the rotational and translational compliance of biomolecules, cells, and tissues.\textsuperscript{[10]} Holonomic control of the handle particles would permit more advanced testing and greater insight into the mechanical properties of biomolecules.

The first demonstrations of combined stable OMT of micro-particles used superparamagnetic nanoparticles dispersed within a polymer matrix.\textsuperscript{[11]} The ferromagnetic nanoparticles were dispersed either randomly or aligned in rows due to coupling of the magnetic dipole moments of each nanoparticle. This type of particle was recently incorporated into functional liquid crystal (LC) structures wherein OT initially positioned the particle within the LC matrix, and an external magentic field was later used to deform the LC by rotating and displacing the particle.\textsuperscript{[12]} Since the properties of LCs may be influenced by sufficiently strong magnetic fields, it is necessary for the inclusion particle to have a magnetic response strong enough to allow particle manipulation without inducing a direct response of the LCs director. There is, however, a limit to the magnetic response that can be obtained from such a particle. Increasing the volume of magnetic material within the matrix will generate a larger magnetic moment and enable increased MT performance. Unfortunately, the added concentration of magnetic particles will also lead to increased optical scattering and absorption of light from the ferromagnetic inclusions distributed throughout the bulk of the sample. Increasing the optical scattering and absorption from additional nanoparticles will lower the stability of any optical trap. Stable OT will be prevented altogether above a particular threshold concentration of the nanoparticles within the particle’s matrix.

Combined OMT was also demonstrated to some extent using Janus particles. These particles are formed by depositing a thin film of ferromagnetic material onto a microscaled particle. Janus particles, in which half of the exposed particle surface is covered with an optically opaque magnetic layer, have been optically trapped in an annular region. This annular trapping is characterized by containment of the trapped particle within a circular region around the optical axis.\textsuperscript{[13]} Janus particles were also observed to spin in place, due to absorbed angular momentum from the trapping laser. The combination of spinning and annular trapping prevent the robust translation of magnetic Janus particles using OT. Patchy particles, in which the metalized portion of the sphere is less than 50% of the overall surface area, have generated a more localized and stable OT. “Dot-Janus” particles were formed by partially masking the particles by first submerging a monolayer into photoresist so that only a cap portion of the particle was exposed. The patch was then formed by evaporating cobalt onto the exposed particle and photoresist surface, then dissolving away the photoresist and releasing the particles from the metal film and substrate via sonication.\textsuperscript{[13]} The particles created by this process are characterized by the localization of ferromagnetic material to a spherical cap covering approximately 20% of the particle’s surface. OMT was demonstrated by stably trapping the dot-Janus particles along the optical axis. Due to the large volume of magnetic material that could be deposited, dot-Janus particles demonstrated the largest magnetic response of any optically trappable particle available to date. Unfortunately, widespread use of dot-Janus particles has been hindered due to robustness issues with the synthesis procedure. The manufacturing parameters are critically sensitive to the diameter of the base particle since the capillary effects involved with particle immersion into the photoresist are highly dependent upon the particle size. Tight tolerances of the base particle are therefore required in order to generate repeatable batches of dot-Janus particles. The sonication process used to release the dot-Janus particles from the substrate relies on the presence of a stress concentration at the particle-photoresist interface. This stress concentration becomes smaller with decreasing areas of dot patches. This decreasing stress concentration leads to a trade off between patch area and patch thickness which resulted in an effective limitation to the amount of ferromagnetic material that could be applied.

The current availability of bifunctional particles capable of OMT is a limiting factor for several applications. Particles containing ferromagnetic nanoparticle inclusions are commercially available, but these particles are limited in the range of magnetic forces that may be applied without jeopardizing the capability of OT. Patchy particles have shown promise in delivering greater ranges of magnetic control, but current manufacturing processes for these particles are difficult to control and are not amenable to large batch processing. The motivation for this research is to demonstrate a means of fabricating multifunctional particles capable of OMT that alleviate these barriers. The volume of ferromagnetic material, and therefore the upper limit of magnetic force, permitted with GLAD surpasses that of any previous optically trappable particle. Furthermore, the fabrication process for creating magnetic GLAD particles is robust and multiple batches may be fabricated in parallel. The synthesis procedure also enables robust control over the size of the ferromagnetic patches, both in terms of thickness and surface area, over a wide range of particle sizes. Once developed, the magnetic GLAD particles described here will have numerous applications including as a handle particle for force spectroscopy experiments or as functional inclusions within LC systems.

2. Optical Trapping of Patchy Particles

The physical explanation of OT with patchy particles may be described using a geometric ray tracing approach.\textsuperscript{[14]} For particles in the Mie regime, where the particle diameter is large with respect to the trapping wavelength of the laser, the incident field may be modeled as a series of rays imparted with an energy proportional to its local field. Each of these modeled rays therefore carries momentum proportional to this imparted energy. When the rays strike the surface, they are reflected and refracted according to Snell’s law with energy distributed to each new ray according to Fresnel’s equations. The ray will then strike an opposing surface of the sphere where again the power will be distributed to a reflected and refracted ray. Since the direction of the ray changes at each surface interface, the optical momentum carried by the ray also changes directions. An optical force must therefore be applied to the particle in order to conserve momentum. The total optical force applied to the particle can be approximated by summing the net force imparted from each ray over every
reflection. Stable trapping occurs at a location where the net force is locally zero and the force imparted due to slight disturbances in particle positions leads to forces acting in the direction of the stable position.

For simple dielectrics, such as in Figure 1a, nearly all of the energy from each incident ray is transmitted at every surface interface. The radial momentum terms are evenly balanced generating a radial trapping force and a stable position on the optical axis. Due to scattering caused by an index mismatch between the particle and surrounding fluid, the stable optical trapping position generally lies slightly above the focal plane when the optical path is transmitted from below the particle. At this axial position, the slight focusing of the rays balances the optical momentum lost in the axial direction due to reflections. In the case of a Janus particle with 50% of the surface area covered by a metallic film (Figure 1b), the light incident upon the metallic portion of the particle leads to a scattering force due to its absorption and reflection. This prevents the Janus particle from ever being stably trapped in three dimensions. Additionally, the uneven absorption of light in the radial direction leads to a net optical torque on the particle inducing on-axis spin. Patchy particles (Figure 1c) can be stably trapped. This trapping arrangement requires that the metallic patch be aligned perpendicular to the optical axis to prevent an optical sail effect in which the patch, aligned with the optical beam path, catches the majority of the rays and is driven away from the focal position due to the increased absorption and reflection.

3. GLAD Particle Fabrication

The high throughput manufacturing and uniformity issues related to the fabrication of dot Janus particles may be overcome by instead generating patchy particles using GLAD. This process relies on the shadowing effect of adjacent particles in a close-packed structure when the substrate normal is angled away from the deposition source as in Figure 2. Similar GLAD procedures have also been employed to develop particles with novel optical properties such as circular dichroism, nanostructures such as columnar nanostructure arrays, and to fabricate magnetic chiral particles capable of “swimming” when exposed to rotating uniform magnetic fields. The standard metal evaporation used in GLAD synthesis is a directional processes in which an uninterrupted line of sight is required between the evaporation source material and the target substrate. When a monolayer of particles is tilted, each particle casts a shadow onto the particles behind it, effectively masking the particles and preventing complete coverage of the monolayer substrate. This shadowing effect can be taken advantage of to control the surface coverage of the deposited material which is crucial to enabling the OT capability of the generated particles. The actual metal coverage pattern and surface coverage depends not only on the tilt of the substrate with respect to the evaporation source, but also on the in-plane rotation of the close packed particles. Decreasing the substrate tilt leads to a higher surface coverage as a lesser portion of each particle is masked. Changing the in-plane rotation will yield different patterns of metallic patches as shown in Figure 3. Certain monolayer formation techniques such as Langmuir–Blodgett (LB) troughs are capable of generating extremely large regions of uniformly oriented grains potentially allowing for precise control of both tilt and rotation. However, LB techniques require significant infrastructure investment which can be prohibitively expensive for non-high-volume production level synthesis. Spin coating techniques in which particles from a fluid suspension are deposited onto a substrate are more common and less expensive techniques for low to mid-volume production synthesis for generating monolayers, even though they suffer from material loss issues and are subject to edge bead effects. Spin-coating enables large regions of close-packed structures to be formed in multiple grain regions. The rotation indices of these grains are
The calculated patterns one would expect from each of these alignments.

a–d) SEM images of non-magnetic GLAD particles along with the calculated deposited patterns. Monolayers of silica particles, created using the same process described in the Experimental Section, had a thin film of Au sputtered onto their surface to create Janus particles and then a 100 nm thick layer of Ti was evaporated onto the surface using the GLAD technique. These metals were chosen to provide adequate contrast in the SEM while preventing distortions to the image caused from scanning the electron beam in the presence of magnetic materials. The monolayer formation produced multiple close-packed grains which resulted in different metalization patterns. a–d) SEM images in which the grains were rotated approximately 3°, 10°, 20°, and 27°, respectively with an average 82° substrate tilt. Scale bars in the SEM images represent 1 µm.

distributed uniformly across the substrate; therefore, multiple orientations of the particles must be considered when predicting the coverage of the patchy particles.

The area of metal coverage on a particle due to the GLAD technique can be predicted from a parametric analysis. First, assume that the target particle on which the metal is being deposited is located at the origin of a Cartesian coordinate system \((x_0, y_0, z_0)\) and another particle which is casting a shadow on the target particle is located at some point \((x_p, y_p, z_p)\) with respect to this origin. The source material that is being evaporated is also assumed to be located on the \(z\)-axis at a distance \(z_e\) from the target bead. The shadow cast by a particle located at \((x_p, y_p, z_p)\) may be modeled by parametric Equation (1)

\[
\begin{align*}
\Gamma(\alpha, \beta) &= x_e(\alpha, \beta) z_e - \rho \sin \alpha \cos \beta \frac{z_e - z_0}{z_e - z_0} + y_e(\alpha, \beta) \frac{z_e - \rho \sin \alpha \cos \beta}{z_e - z_0} + \rho \sin \alpha \cos \beta \hat{k} \\
&= x_e(\alpha, \beta) \frac{\rho \sin \sin (z_e - z_0)}{z_e - \rho \sin \alpha \cos \beta} + y_e(\alpha, \beta) \frac{\rho \sin \sin (z_e - z_0)}{z_e - \rho \sin \alpha \cos \beta} + \rho \sin \alpha \cos \beta \hat{k} \\
x_e(\alpha, \beta) &= \frac{\rho \sin \sin (z_e - z_0)}{z_e - \rho \sin \alpha \cos \beta} \frac{x}{\sqrt{\rho^2 - (x_e(\alpha, \beta) - x_0)^2}} \\
y_e(\alpha, \beta) &= \frac{\rho \sin \sin (z_e - z_0)}{z_e - \rho \sin \alpha \cos \beta} \frac{y}{\sqrt{\rho^2 - (x_e(\alpha, \beta) - x_0)^2}} \\
\end{align*}
\]

where \(\rho\) is the radius of the particle. The variables \(\alpha\) and \(\beta\) represent a spherical coordinate system sharing an origin with the target particle and defined by the equations:

\[
\begin{align*}
x &= \sin \alpha \cos \beta, \\
y &= \cos \alpha, \\
z &= \sin \alpha \cos \beta.
\end{align*}
\]

In most metal evaporation systems, the separation distance between the evaporation source and the substrate (on the order of tens to hundreds of mm) is much greater than the separation distance between shadowed particles (on the order of several micrometers). In this limit, the above equation can be greatly reduced to the form presented in Equation (2) as

\[
\Gamma(\alpha, \beta) = (\rho \sin \alpha \sin \beta) \hat{i} \\
+ \left[ y_0 + \sqrt{\rho^2 - (\rho \sin \beta - y_0)^2} \right] \hat{j} \\
+ (\rho \sin \alpha \cos \beta) \hat{k}
\]

Using the above equation, the intersection of a shadow cast from adjacent particles in a close-packed formation onto a target particle can be calculated and the subtended area of an exposed particle may be determined. A parametric analysis was performed assuming that each substrate contained grains with uniformly distributed grain alignments. The results (Figure 4a) demonstrate that by controlling the tilt angle of the substrate, the metal surface coverage may be easily modulated with less than a 1% standard deviation for metalized surface areas less than 15%.

Alternatively, GLAD can be controlled by aligning the substrate vertically and shifting it off center as demonstrated in Figure 2b. This arrangement eliminates the need of customized tooling used to hold the substrate at a fixed angle. A single vertical mounting fixture can be created and the degree of metalization can be adjusted based on the mounting offset distance. By aligning multiple vertical substrates radially around the evaporation source, this arrangement allows for multiple substrates to be exposed simultaneously with unique surface coverage settings. The disadvantage of the offset alignment is an increased variability of surface coverage due to effective tilt angle changes from the bottom to the top of the substrate. However, for most applications, this increased variability will be sufficiently small due to the small angle assumption described above. The expected surface coverage distribution (Figure 4b) for a 1 in. tall substrate was calculated based upon the substrate offset distance using the previously mentioned parametric approach. Even with the increased variation from the offsetting alignment, the standard deviation for the metalization areas remained less than 1%, indicating that offsetting the substrate is a viable method for reliably controlling the degree of surface metalization.
The metalized surface area results described above are independent of the actual particle diameter, assuming that particle diameters within the monolayer are well controlled and of similar size. Monolayers formed with larger or smaller uniform monolayers will yield the same percent surface coverage using the same tilt orientation. Magnetic GLAD particles can therefore be generated using base particles from separate batches without the need to modify the synthesis parameters. Unique magnetic GLAD particles designed with a size specific to an application may therefore be synthesized without extensive testing or production overhead, as opposed to current synthesis methodologies for OMT patchy particles. Higher metal surface coverage rates could be obtained by further reducing the tilt and exposing larger regions of the bead to the metal evaporation source; however, this would lead to poorer performance during OT and is not explored in greater detail here.

4. Experimental Section

As a proof of concept, magnetic GLAD particles were synthesized with 10% mean surface coverage. First, a colloidal solution of either silica or polystyrene (PS) particles with nominal diameters of 3.17 and 3.11 µm respectively (Bangs Laboratories, Inc.) were washed by centrifugation and resuspended in ethanol at a concentration of 20% wt/wt. One inch square glass substrates were cleaned by subsequent immersion into acetone, isopropyl alcohol, deionized water, and finally piranha solution. This cleaning process was intended to remove any biological contaminants and increase the hydrophilicity of the surface to aid with the monolayer formation. The glass substrate was then dehydration baked on a hotplate at 200 °C for at least 5 min immediately before use. Next, 200 µL of the washed colloidal solution was pipetted onto the substrate. This solution was allowed to stand on the substrate for either 30 s (silica solutions) or 120 s (PS solutions) before spinning at 4000 rpm for 30 s. This spinning process produced large areas of colloidal monolayers on the surface that were suitable for GLAD. Edge beads, or areas near the substrate edge in which multilayer or unordered colloids formed, were manually removed from the substrate after spinning. Once the colloidal monolayer substrates were created, the substrates were mounted vertically onto the evaporator platen with a 46 mm offset from center using a custom designed fixture. The evaporator used in this study (PVD-75, Kurt J. Lesker Co.) has a separation of 300 mm from the evaporation crucible and the platen plane which results in a mean 10% surface metalization at the given offset as predicted in the previous section. A thin adhesive layer of either Cr or Ti was first evaporated onto the substrate followed by a layer of Co with desired thickness to provide the ferromagnetic properties. No adjustments were made to the substrate position in between the depositions. If the synthesized magnetic GLAD particles were intended for a potentially corrosive environment, or if a metallic non-cobalt surface was desired for chemical functionality, a subsequent thin film with the desired chemical properties could also be deposited at this stage. However, such an additional layer was not deposited in any of the present demonstrations. Once the evaporation process was complete, the particles were lifted off of the substrate by sonicating for 5 s in a 0.05% Liquinox solution. SEM images of representative particles were obtained (Figure 3) showing the actual deposition patterns. Small patches of metalized areas that accurately match the predicted patterns expected from the theoretical parametric analysis are clearly visible.

Optical transmission and reflection properties of the metallic patch region vary based upon the choice of patch composition. Figure 5 shows the calculated Fresnel power coefficients for the specific case of a 5.0 nm Ti and 50.0 nm Co patch deposited onto a PS particle and suspended in water. These coefficients describe how the power of an incident ray is distributed between reflected and transmitted rays. In general, this metallic patch has a larger proportion of power transmitted to the reflected ray or lost to absorption when propagating into the particle compared to the PS and water interface. When light propagates out of the particle, nearly all of the power at a patch interface is lost to absorption, regardless of the contact angle.

In order to test the optical and magnetic trapping responsiveness of these particles, a custom OMT apparatus was designed. Optical trapping was performed using a continuous wave 1064 nm wavelength infrared laser (Laser Quantum, Ventus IR 3W) with optical powers ranging from 2 to 10 mW measured at the objective port. The OT was formed using a high numerical aperture (100x/1.3NA, Zeiss Plan-Neofluar) mounted in an inverted microscope (Zeiss, Axiovert). The optical train also included a fast scanning mirror (FSM) (Newport, FSM-300) and a spatial light modulator (SLM) (Holoeyley, PLUTO) to dynamically control the OT location. The SLM is capable of encoding phase masks into the propagating beam which, after propagating through the focusing objective, result in the holographic projection of multiple OTs with arbitrary 3D distributions.[19,20] For the purposes of this study, the OT generated by the OT consisted only of single traps that varied in locations along the optical axis. The FSM is used to dynamically control the position of the OT within the focal plane of the OT. This is done by applying tip and tilt to the wavefront which laterally translates the OT proportional to the angular offset of the FSM.[21] Both the FSM and the SLM were located in conjugate planes to the Fourier plane of the objective. Lenses in a 4-f configuration...
index of refraction of 1.5717 for PS, [27] 1.333 for water, (3.4613+3.3135i) for Co. [29] These properties were selected assuming an incident wavelength of 1064 nm.

The coefficients for the multilayer patches were calculated using a recursive method [26] using and posed of 5.0 nm of Ti and 50.0 nm of Co deposited onto a PS particle and suspended in water. Figure 5. Calculated Fresnel reflection (R) and transmission (T) coefficients for patches composed of 5.0 nm of Ti and 50.0 nm of Co deposited onto a PS particle and suspended in water. The coefficients for the multilayer patches were calculated using a recursive method [26] using and index of refraction of 1.5717 for PS, [27] 1.333 for water, (3.4613+3.3135i) for Ti, [28] and (2.9031+5.7923i) for Co. [28] These properties were selected assuming an incident wavelength of 1064 nm.

were used to separate the SLM, FSM, and the objective’s Fourier plane.

The magnetic trapping system (Figure 6) incorporated four mu-metal pole tips with electromagnetic coils mounted on the end of the tip away from the sample. Each of the coils was formed from 80 turns of 24 AWG motor winding wire around an iron core. An iron yoke connecting the four coils was also used to reduce the magnetic resistance of the system. Each coil was independently driven by a high power op-amp (Kepco, BOP-20–5M). When equal and opposite current densities are applied to opposing coils, an approximately uniform magnetic field is generated in the central trapping region between the pole tips due to the pole tip geometry. [11a] A controlled magnetic flux density between ±5 mT, measured with a Gaussmeter (LakeShore 455) could be applied from a single pair of mu-metal poles tips. A microfluidic chamber was also designed to align the pole tips and deliver the magnetic particles to the optical trapping region. This entire magnetic trapping sub-system, excluding the operational amplifiers, was mounted onto a custom designed microscope stage used to adjust the alignment between the optical and magnetic systems.

Due to the large ferromagnetic shape anisotropy, the magnetic moment, \( \mathbf{m_p} \), is effectively fixed in the same plane as the metallic film. When subjected to an external magnetic field, \( \mathbf{H}_{ext} \), a torque is applied aligning the particle’s magnetic moment to the external field according to \( \tau = \mu_0 \mathbf{m_p} \times \mathbf{H}_{ext} \), where \( \mu_0 \) is the magnetic permeability of vacuum. This torque aligns the magnetic moment of the particle with the external field and constrains one of the rotational degrees of freedom (DOF). Rotating the direction of the applied magnetic field correspondingly rotates this constrained DOF. When an OT is applied in conjunction with the MT, the remaining five degrees of freedom will be constrained. The OT applies a trapping force that constrains the three translational DOF of the particle. The location of the OT may be translated using the fast scanning mirror or dynamic holograms as described above permitting these translational DOF to be manipulated. Additionally, the shape anisotropy of the metallic patch constrains the remaining two rotational DOF, preventing the patch from rotating into the optical path. Overall, this system enables control of the magnetic GLAD particles in four DOF while constraining the remaining two. Additional control of the remaining two DOF could be obtained by incorporating additional magnetic actuators and properly calibrating the magnetic and optical characteristics of a particle, but was not pursued in this study.

Figure 6. CAD image of the microfluidic optical and magnetic trapping apparatus. A PDMS microfluidic chamber flows particles, shown red, into the trapping region. The PDMS mold also serves to align four mu-metal poles with the optical trapping regions. An iron core solenoid with 70 turns of 20 AWG motor winding wire is positioned atop the end of each pole tip. The iron yoke connecting each solenoid is used to reduce the magnetic impedance of the system. The high NA microscope objective may be seen through the PDMS microfluidic chamber and coverslip.

Figure 7 shows a composite image of a magnetic GLAD particle translationally fixed with an OT and rotated around the optical axis by adjusting the angle of the applied magnetic field, demonstrating the ability for simultaneous OMT using magnetic GLAD particles. This particle was optically trapped approximately 15 µm above the coverslip surface demonstrating that the OT is in fact capable of full 3D trapping and manipulation of magnetic GLAD particle both translationally and axially. As the orientation of the magnetic field was rotated around
the optical axis, the optically trapped magnetic GLAD particle rotated accordingly to align with this field. In order to accentuate the magnetic patch in these optical microscopy images, a dynamic hologram was applied with the SLM to translate the optical trap out of the focal plane of the microscope so that diffraction effects made the patch more optically discernible. This axial adjustment also demonstrates the ability to successfully trap and manipulate magnetic GLAD particles in all three translational dimensions. Particle rotations due to magnetic actuation without the presence of an OT were also observed indicating that the rotations could be controlled using the MT system independently.

Additionally, oscillating currents perfectly out of phase with one another were applied to each pair of electromagnets generating a rotating magnetic field in the trapping region. The rotating field enabled rotation of the magnetic GLAD particle which was phase locked with the driving current. Phase locked rotations were observed in both clockwise and counterclockwise directions at frequencies up to 10 Hz, which was the upper limit achievable due to the time constant of the electromagnetic system. Furthermore, simultaneous translational and rotational control of these particles were demonstrated by translating the OT position using a fast scanning mirror while simultaneously rotating the magnetic field to spin the particle. A video demonstrating this simultaneous optical translation with magnetic rotation is provided in the Supporting Information. Particle ejections due to thermal vibration of the particles were not observed during the course of this study providing a distinct advantage over previously reported metalized Janus particles.

Next, the OT ejection force was measured for a series of magnetic GLAD particles to quantify the effects of the increased scattering and absorption from the metallic patch. In this test, the FSM translated a trapped particle in a saw-tooth pattern with constant velocity. The relative velocity of the particle with respect to the surrounding fluid induces a viscous drag force displacing the particle within the trap. When this drag force is great enough to overcome the stiffness of the OT, the particle will be ejected. The force resulting in an ejection of the particle may be used to quantify the trapping efficiency of an OT. In this study, a series of particles were trapped by an OT and translated to a position of 18 µm above the coverslip surface. The FSM then translated the particles in a saw-tooth pattern with a displacement amplitude of 20 µm centered on the optical axis. The velocity of the particle was increased by 1 µm s\(^{-1}\) in each subsequent oscillation of the saw-tooth displacement. After the particle was ejected, the scattering force in the axial direction caused the particle to eject along the optical axis and out of the field of view of the microscope. Viscous drag forces on a spherical particle were calculated based upon the displacement velocity, fluid viscosity, particle radius, and offset height from the trapping surface. Ejection forces at several power settings were obtained for magnetic GLAD particles synthesized from PS with a 5.0 nm Ti adhesive film and a 50.0 nm Co patch along with a control group consisting of only PS particle. The escape forces increase linearly with applied optical power and follow a similar trend for each test grouping.
Finally, the magnetic properties of the magnetic GLAD particles were examined using an alternating gradient magnetometer (AGM) (PMC MicroMag 2900, Lake Shore Cryotronics). Solutions of suspended particles with 5.0 nm Ti and 50.0 nm Co patches at a concentration of 98.90 ± 4.89 particles per µL were prepared and verified using standard optical microscopy techniques. Magnemometer samples were then prepared by depositing 2 µL of solution onto a 4 mm square section of coverslip. The solution was allowed to evaporate, depositing magnetic GLAD particles onto the coverslip. Hysteresis curves for these samples are shown in Figure 9. The hysteresis parameters determined from this analysis include a magnetic remanence \( M_r \) of 358.1 nemu, a saturation magnetization \( M_s \) of 1041 nemu, a coercivity of remanence \( H_{r\text{cr}} \) of 247.5 Oe, and a coercivity \( H_{c\text{r}} \) of 165.6 Oe. Since the saturation magnetization is additive, the average magnetic saturation for a single magnetic GLAD particle with the described composition is determined to be approximately 5 pemu per particle. Ratios of the hysteresis parameters along with the shape of the hysteresis curve are also useful for providing a grain size independent method to determine the magnetic domain state. The measured hysteresis parameters, \( M_r/M_s = 0.3440 \) and \( H_{r\text{cr}}/H_{c\text{r}} = 1.495 \), is indicative of single domain-like or pseudo-single domain (PSD) magnetic properties which implies that the individual patches are composed of single domains or small multidomain states.

A parameter space analysis was conducted to predict the metallic surface coverage of a GLAD particle as well as the distribution of surface coverages from typical batch manufacturing techniques. The particle manufacturing process greatly improves upon previous OMT particles by increasing the concentration of magnetic material while still enabling stable optical trapping. The volume and surface coverage of ferromagnetic material may be robustly controlled utilizing the GLAD technique to tailor the desired range of magnetic forces and torques generated by the particle in a given system. The adjustment of the magnetic coverage can be accomplished without any dedicated hard tooling and is amenable to manufacturing scale-up.

A custom optical and magnetic trapping system was also demonstrated for decoupled control of the translational and rotational motions of magnetic GLAD particles. Magnetic GLAD particles were manipulated in three translational degrees of freedom utilizing dynamic holograms and a fast scanning mirror to modulate the trapping laser. Magnetic torques were also shown to rotate the particles while the particle was stably optically trapped. These magnetic GLAD particles will have beneficial applications in optical force microscopy where the increased range of forces and decoupled rotation and translation control may be used to interrogate novel molecular and cellular processes. Additionally, the magnetic GLAD particles show promise for several manufacturing applications including the incorporation of magnetically functional colloids into liquid crystal systems as well as handle particles for force spectroscopy experiments.

### Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

### Acknowledgements

Research reported in this publication was supported by the National Institutes of Health under Award No. R221 GM104576. The content is solely the responsibility of the authors and does not necessarily represent the official views of the National Institutes of Health. The authors would also like to thank Richard Bono and Dr. Rory Cottrell from the Paleomagnetic Research Group at the University of Rochester for assistance collecting the magnetic hysteresis data.

Received: March 5, 2015
Revised: April 22, 2015
Published online: May 22, 2015
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