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How to Manipulate Nanoparticles with an Electron Beam?

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Recently discovered vortex electron beams[1–3] share the wave property of helical wavefronts with vortices observed in optical, acoustical and radio waves.[4–8] This vortex property gives the wave orbital angular momentum (OAM) which can be transferred to particles to make them rotate as was demonstrated with light optics and acoustical waves.[9–12] Electron vortex beams also carry an orbital angular momentum which is quantised to $m\hbar$ per electron with m the topological charge of the vortex. Breaking the circular symmetry of the vortex wave in free space by elastic interaction with a nanoparticle that does not have circular symmetry is theoretically expected to lead to an exchange of angular momentum between beam and particle because angular momentum is no longer a good quantum number.[13] This means that also with electron beams one expects that transfer of OAM to nanoparticles is possible. Here, we show experimental evidence of OAM induced rotation of supported Au nanoparticles by impact with $m = \pm 1$ electron vortex beams with the direction of rotation determined by the sign of m. This observation is a qualitative confirmation of the theoretical prediction and adds transfer of angular momentum to nanoparticles to the already wide range of possible applications of electron vortex beams. The observed rotation of the nanoparticles also allows studying the friction of the Au nanoparticles with their support on the nanoscale leading to a rotational diffusion coefficient in agreement with previously reported theoretical values.[14]

Electron vortex beams have been produced in transmission electron microscopes in different ways but they all have in common that the electron wave function in a plane perpendicular to the optical axis can be described by a product of azimuthal and radial parts:

$$\Psi(r, \varphi) \propto e^{im\varphi} f(r, z) \tag{1}$$

With m the topological charge and f(r,z) the radial distribution function that generally depends on z as the beam focusses and diverges along the optical axis z. Making use of a fork aperture in the condenser plane of a transmission electron microscope as sketched in **Figure 1** we can produce vortex beams of $m = -1,0,+1$ with diameters down to the atomic level[15] depending on the convergence angle α. Neglecting lens aberrations we can obtain an expression for the beam profile at the beam waist:[16]

$$f(r, 0) = \int_0^{\alpha} J_m(kr) k dk \tag{2}$$

In this experiment we make use of dispersed colloidal Au nanoparticles on a supporting thin film of Si_3N_4 of thickness 15 nm. We illuminate one such nanoparticle with a diameter of approximately 3 nm with an electron vortex with a radial distribution matching the size of the nanoparticle as sketched in Figure 1. Imaging the elastically transmitted electrons through the particle allows us to observe the lattice planes of the particle as in conventional high resolution transmission electron microscopy.

Figure 2 shows the resulting images as a function of time for illuminating the particle either with the left or right handed vortex beam $m = +-1$. Arrows in the images indicate the (111) lattice planes in the Au particle clearly rotating as time evolves over the course of minutes. A more detailed figure with diffractograms calculated by Fourier transforming the images is given in supplementary information.

Most notably, the rotation direction depends on the sign of m. This is a strong indication, that from all possible electron/particle interactions, the phase of the beam, i.e., the vortex character is crucial here. It must be noted that upon illuminating the particles also slightly deform under the beam. This is common in electron microscopy and also occurs with non-vortex beams on Au particles due to kinetic energy transfer to surface atoms making them migrate. Note that heating of the particles due to inelastic scattering is negligible for the low current densities used here and can be approximated in the mK range.[17,18] The defining character of this experiment is however that even though the particle undergoes some unpredictable surface changes, there is a clear trend in the rotation of an

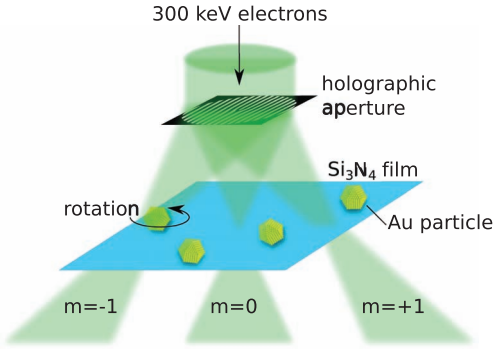

Figure 1. Sketch of the setup to illuminate supported Au nanoparticles with a vortex electron beam making use of a holographic mask with an edge dislocation.

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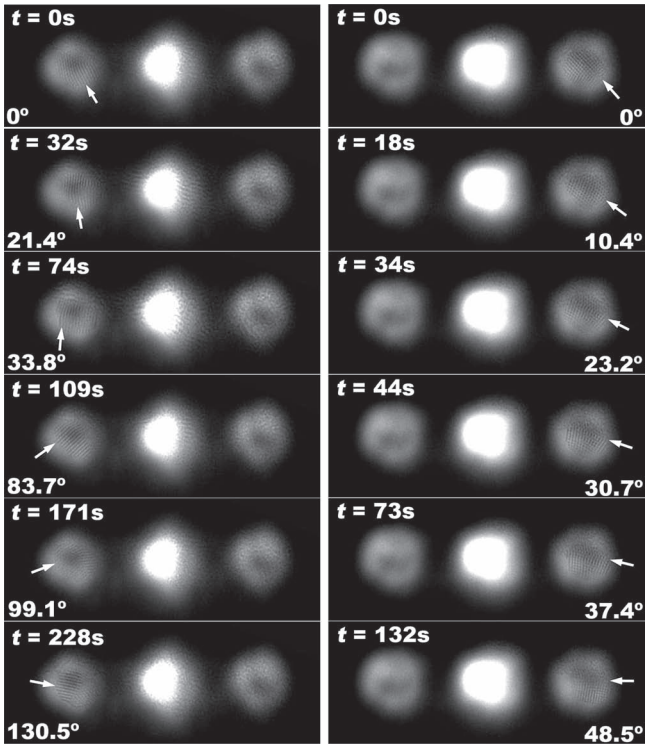

Figure 2. Time lapse series of experimental atomic resolution images of a single supported 3 nm Au particle illuminated with either a $m = -1$ (left) or $m = +1$ (right) vortex beam demonstrating that the particle rotates with time in a direction determined by the topological charge of the vortex beam.

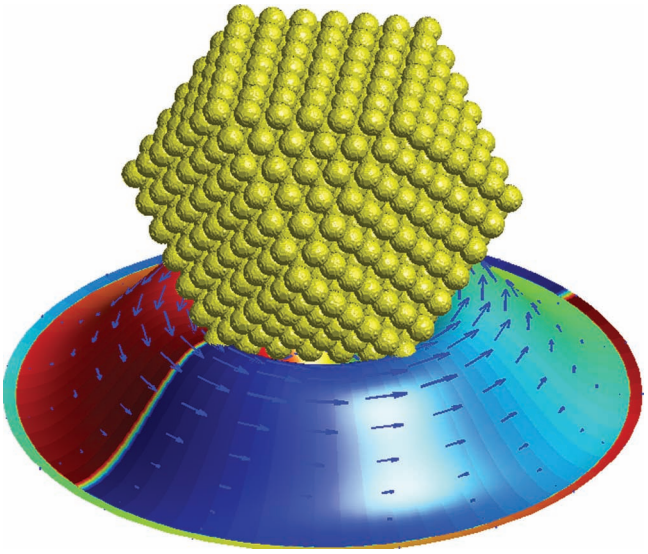

Figure 3. Sketch of the radiation force on a cubeoctahedron Au particle of approximately 3 nm diameter. The color coding of the electron wave shows the phase while the height of the surface shows the intensity. The blue arrows indicate the in-plane probability current which gets partially transferred as a mechanical rotation of the Au particle.

individual particle around the vortex axis depending on the sign of the vortex.

In order to understand the nature of this interaction we sketch the situation in **Figure 3** showing the intensity, phase and rotating transverse current with respect to a model cubeoctahedron Au particle. The interaction can be understood from transfer of angular momentum via elastic scattering and is essentially a QM effect.

A rotational force is transferred via the radiation force as the gradient of the phase has an azimuthal component around the center of the particle. The reason why the particle exchanges angular momentum with the crystal is due to the breaking of cylinder symmetry as the particle has a non-spherical shape and consists of individual atoms. Indeed the symmetry breaking causes the angular momentum operator of the fast electrons to no longer commute with the Hamiltonian describing the interaction with the nanoparticle. The exact interaction with the particle depends on the complicated elastic scattering of the fast electron wave with the electrostatic potential of the electron density of the particle. This is commonly simulated solving the Schrödinger equation making use of either Bloch wave or multislice methods and existing codes can be adapted to take a vortex probe as input.[19] Doing this we calculate the expectation value for $\langle Lz \rangle$ after transmission through the crystal. This loss of angular momentum is transferred to the particle. The exact amount of transferred OAM depends on the size of the vortex, the convergence angle, the exact details of the particle but changing many of these parameters we find the exchange to be approximately 0.1 ħ per incoming electron with a kinetic energy of 300 keV [see supplementary info for simulation details]. Note that also inelastic scattering can transfer angular momentum even when the beam electron is some distance away from the particle as described theoretically by de Abajo et al.[20] which was used to explain experimental observations of an electron tweezer effect.[21,22] As both inelastic and elastic effects occur simultaneously we can regard the elastic calculation as a lower limit for p, the average transfer of angular momentum per electron. The smaller the particles however, the lower the probability for inelastic scattering and therefore we expect the elastic scattering to be dominant. Assuming $p = 0.1$, a homogeneously round particle for the moment of inertia and a typical beam current of 50 pA we obtain a linearly increasing velocity that already reaches 10^{14} rad s^{-1} after a 1 s exposure. In reality, frictional forces between particle and support dissipate a large part of this rotational energy and lower the rotational velocity by several orders of magnitude. Assuming viscous drag we obtain a stationary angular drift velocity under an applied torque τ given as:

$$\varpi_d = \frac{\tau}{f_r} \tag{3}$$

With $f_r = k_B T/D_0$ the viscous friction constant and D_0 the rotational diffusion coefficient.[23] The torque applied by the electron vortex on the particle is given as:

$$\tau = \frac{dL}{dt} \tag{4}$$

with L the total supplied angular momentum to the particle by the beam which rises linearly as $L = pI/et\hbar$ with I the current in the beam, p the average transferred fraction of $\hbar$ per electron and t the time. Inserting this in the expression for the rotational drift velocity and using a thermally activated expression for the diffusion coefficient proposed we get:[23,24,14]

$$\varpi_d = p\hbar\frac{I}{e}D_0\frac{e^{-\frac{E_0}{k_BT}}}{k_BT} \quad (5)$$

For our experimental parameters T = 300 K, p = 0.1 and I = 50 pA and using the theoretical values of Guerra et al. for a situation of smaller Au particles (36 atoms) on a graphite surface (D_0 = 0.77E14 rad^2 s^{-1}, E_{0rot} = 0.509 eV) as the closest match in literature to the present situation we get a rotational drift velocity of 0.32 rad s^{-1} in a similar range as observed in our experiment. The lower drift velocity of approximately 0.01 rad s^{-1} as observed in our experiments can be explained by the higher friction coefficient that is expected for larger particles. Indeed, linearly extrapolating the friction data from a particle consisting of 36 atoms to a particle with 923 atoms assuming that friction scales with the surface of the particle leads to an estimate of 0.037 rad s^{-1} closer to the observed value. The lack of experimental nanofriction data in the literature demonstrates that electron vortex beams could become a valuable tool of investigation in this field, complementing the possibilities of rotating nanoparticles with atomic force microscopy.[25]

Several strategies can now be proposed to lower the friction in order the make the particles rotate at a higher velocity. Increasing the temperature to 400 K would already increase the speed by two orders of magnitude but is difficult to control the temperature at the location of the particles due to the very limited heat transfer through the supporting thin film. Another approach would be to replace the supporting film by a liquid in a so-called environmental cell. Recently promising experimental results were shown on liquid pockets encapsulated between graphene layers[26] and for (non vortex) electron tweezers acting on Al particles in a molten eutectic. Another factor to influence is the shape of the particles; in order to increase the breaking of circular symmetry one could envision that elongated nanoparticles would lead to a higher fraction of angular momentum to be exchanged; the gain via this strategy is however limited to at maximum one order of magnitude. Increasing the topological charge in the beam as in[27,2] would also increase the transferred momentum. The vortex beams used here carry OAM parallel to their propagation direction. In general, however, it is possible to create spatiotemporal vortex beams with an arbitrary angle between OAM and linear momentum.[28] Using such beams could allow imparting orbital angular momentum around an axis parallel to the supporting film setting particles into a rolling rather than sliding motion.

The above described experiments conclusively show that angular momentum can be transferred to nanoparticles setting them into a rotating motion. This observation complements similar observations made with optical and acoustical vortex waves but transposes the possibilities into the realm of nanoparticles and atoms owing to the much smaller dimensions of electron vortex beams. Apart from being of fundamental interest to study friction and mechanical conservation laws on the nanoscale, these observations could be useful to manipulate and rotate nanoparticles inside an electron microscope or to mix fluids on scales approaching 10^{-30} l. Reducing friction could allow experimenters to impart extremely high rotational velocities on nanoparticles and to study at what point they would yield. Orienting and rotating proteins in liquids could be another topic of interest building on recent successes with X-ray diffraction experiments.[29] In atom traps, one could envision imparting angular momentum to the trapped atoms making use of a standing wave of two coherent vortex electron beams of opposite propagation direction. This way the trapped atom can gain in principle an infinite amount of rotational energy possibly leading to interesting physical effects.

Supporting Information

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

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