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Attosecond Forces Imposed by Swift Electrons on Nanometer-Sized Metal Particles

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In previous work, we have explored the behavior of gold nanoparticles (NP) in the aberration corrected STEM, experimentally [1, 2] and theoretically [3], suggesting that nanoparticle movement in the microscope can arise from forces driven by plasmonic fields excited by the passing electron beam.

Looking in more detail, many different kinds of behavior emerge. These include 1) coalescence, discussed previously [1], 2) separation, wherein nanoparticles are driven apart, 3) pushing or pulling of single nanoparticles, 4) atomic level directing of atoms, and 5) complex multiple particle behaviors. Theoretically, many different geometric combinations still need to be tested, even including composite structures: spheres embedded in dielectrics, in contact with conducting layers, near nanopores or protrusions. For instance, using theoretical modeling of electromagnetic fields in the frequency domain, it was found that plasmon-driven forces contribute to the pull and push behavior of single metal NP's during a close approach by the electron beam [3]. However, while this numerical work contributes to our understating of the phenomenon, we still lack a complete understanding of the physical mechanism that causes the intriguing repulsion between single metal NP's and the electron beam.

In this work, we modeled theoretically the time-varying electromagnetic forces acting on a spherical nanometer-sized particle, imposed by a relativistic electron travelling in a non-intersecting geometry (Fig. 2A). Working within the framework provided by Maxwell's equations and special relativity, we identified two fundamentally different mechanisms that produce forces imposed on NP's. At the first stages of the process – corresponding to the time when the electron passes nearby the NP - the NP is subjected to attosecond impulse forces (Fig. 1A), which are generated by the moving external fields acting on the induced charge and current densities. Those forces exhibit both attractive and repulsive natures which are mainly driven by the total electric and magnetic fields, respectively. Figure 1A shows the electromagnetic forces acting a 1 nm aluminum NP, driven by a 120 KV electron travelling with a 0.5 nm impact parameter. Later, femtosecond damped oscillatory reaction forces are generated in the subsequent stages of the process (Fig. 1B). These oscillatory forces are enabled by the well-known surface plasmon oscillations in the NP, triggered by the external excitation. Note that the impulse and oscillatory forces occur during different time scales of the interaction, while the largest contribution to the total linear momentum transfer is generated during the attosecond time scale.

We have also mapped both the total electric and magnetic fields over the NP surface as a function of time to get insight into the origin of the forces. Our analysis shows that the attosecond impulse forces result from a large field gradient across the NP during the close passage of the swift electron. Figure 2B shows the high concentration of total fields during the passage. The electric, attractive forces have dielectric nature, where the induced dipole moments in the NP system are subjected to attractive forces. On the other hand, the magnetic, repulsive forces have a diamagnetic nature, where circulating currents induce magnetic moments pointing in directions opposite to the external magnetic field (Lenz's law). This mechanism generates repulsive forces which compete with the attractive electric forces. During femtosecond time delays, response fields display a damped oscillatory nature, which leads to the generation of smaller, plasmonic forces. We think this force arises from emission of photons during plasmon decay, but verification of this will require understanding photon intensity angular distributions in detail. Our results provide progress in understanding the coupling between relativistic electromagnetic fields and nano-sized volumes of matter, bringing light into the origin of the repulsive forces acting on NP's.

References:
[1] P. E. Batson, et al, *Nano Letters* 11 (2011) 3388.
[2] P. E. Batson, et al, *Ultramicroscopy* 123 (2012) 50.
[3] A. Reyes-Coronado, et al, *Physical Review B* 82 (2010) 235429.
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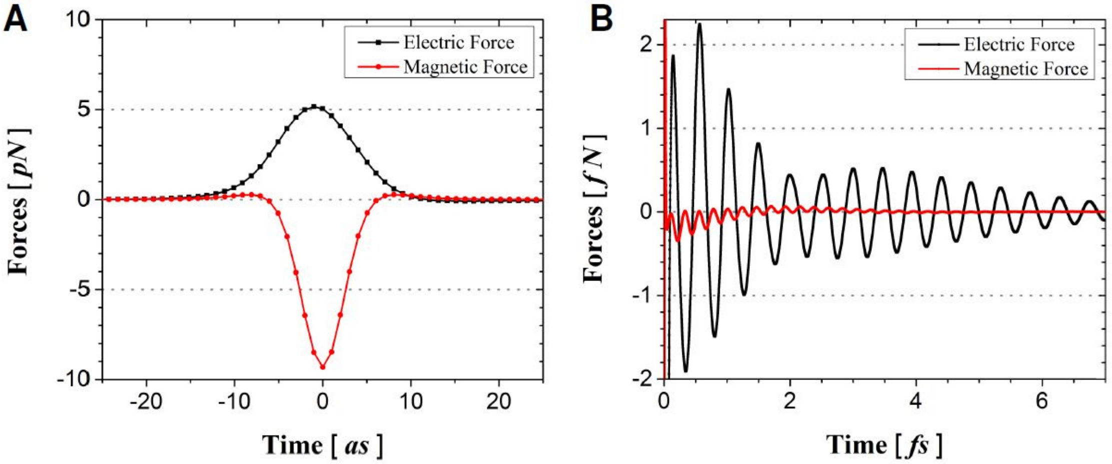

Fig. 1. Resultant time-dependent electric (black) and magnetic forces (red) acting on an Al 1nm nanosphere induced by a 120 KV electron passing with impact parameter of 0.5 nm. (A) Impulse attosecond electromagnetic forces and (B) oscillatory femtosecond forces.

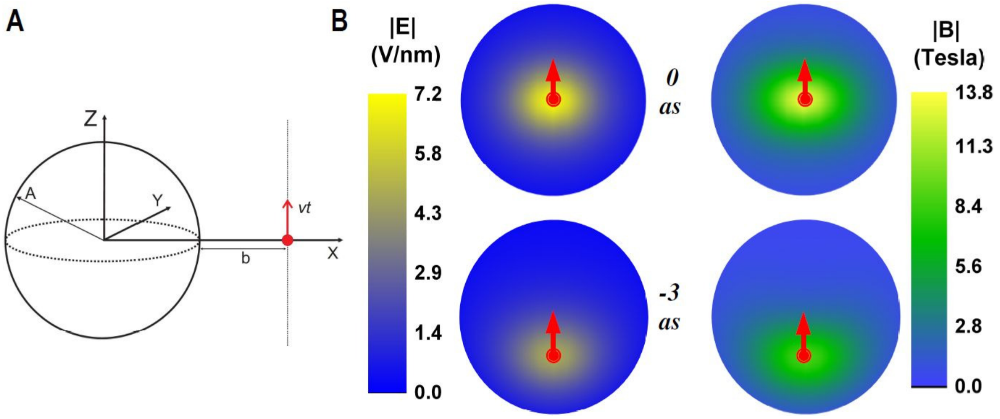

Fig. 2. (A) Schematic Representation of the interaction between an electron and metal NP in an aloof geometry (B) Sequence of 2D projected images of the (left) electric and (right) magnetic fields along the YZ plane. The electron position is indicated by a red dot.

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