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Ultrafast Plasmonic Forces Imposed by Fast Electrons on Metal Particles

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Recently, several groups reported movement of nano-objects apparently driven by electron beams [1-3]; thus pointing out the possibility of using electron-beams for effective manipulation of nanostructures. Among the different types of behavior reported -- such as pushing and pulling of single nanoparticles (NP's), rotation, and NP coalescence -- the observation of both attractive and repulsive forces driving movement of gold NP's has been the most difficult to understand. Attractive forces between an electron and a NP seem entirely reasonable, simply by imagining a positive image charge within the NP, induced by the negative charge on the swift electron [4]. But understanding of a repulsive force still remains elusive. In this work, we have studied the temporal evolution of forces acting on a metal sphere induced by a swift electron; in particular aiming to explore the dynamics of plasmon excitations and their role in the generation of ultra-fast Lorentz forces.

We modeled theoretically the time-varying electromagnetic forces acting on a spherical 1nm-radius Au NP, imposed by a 120KV- relativistic electron travelling in a non-intersecting geometry (Fig.1). We found fascinating results which bring more elements to our understanding of the attractive and repulsive forces in the context of plasmonic response: (i) surface plasmons at optical frequencies (~ 2.5 eV) produce oscillatory forces at femtosecond times which are likely mediated by the emission of photons into the electromagnetic fields during the plasmon decay. At frequencies around 25 eV (deep ultraviolet range), we noticed a second plasmonic instability that apparently originates with the excitation of 5d electrons in gold [5]. Interestingly, these two plasmonic modes occur at quite distinct times, following the passage of the swift electron. We think that these two modes are weakly coupled, leading to formation of a beating pattern in the response forces, in spite of their large energy mismatch. These plasmonic features produce very weak oscillatory forces at moderate distances, so their contribution to the total momentum transfer to the NP is quite small. (ii) At higher energies, we notice a strongly confined surface *wake pattern* on the sphere, having a wavelength that is significantly shorter than the particle diameter. This wake pattern lags the passing electron and is composed of positively charged regions (holes) between large regions of negative charge. It induces, on average, a repulsive force in the spherical NP (Fig. 2). (iii) During the close approach at atto-second times, external electric and magnetic fields imposed by the swift electron interact with induced charges and currents within the sphere to produce strong attractive and repulsive forces. These forces compete one against the other, resulting in a net force which is primarily a dielectric attraction during the approach of the swift electron and a diamagnetic repulsion as the electron leaves. These attosecond forces contribute most of the total momentum transfer from the electron to the NP.

Our results provide progress in understanding the physical origin of the repulsion behavior of NP's driven by swift electrons, pointing out the possibility of wide exploration of ultrafast phenomena in nano-sized systems.

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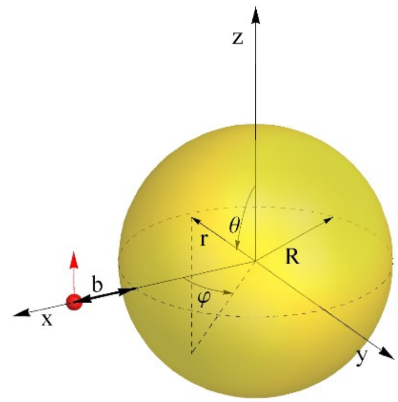

Fig. 1. Modeling geometry for the electromagnetic interaction between an electron and metallic nanosphere in an aloof geometry: the electron travels in the +z direction, with speed v along the trajectory r = (R+b, 0, v*t*), where R is the radius of the sphere and b is the impact parameter.

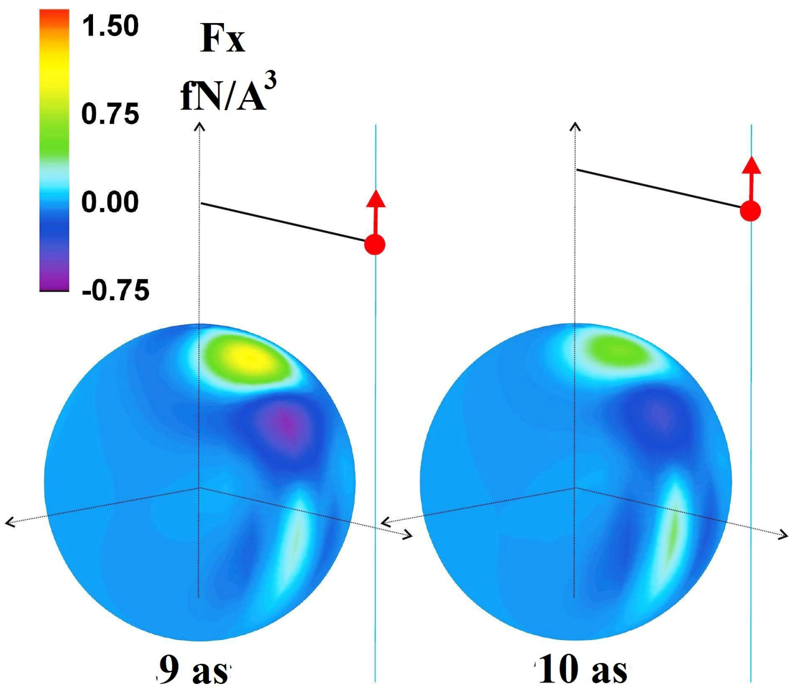

Fig. 2. Lorentz forces resulting from the interaction between an induced surface wake and a swift electron (red dot) during attosecond times. The broad accumulation of negative charge induces repulsive electric forces which lead to weakening of the attractive electric contributions. Note the forces (and the charge density fluctuations) display a cone-like pattern spreading over the surface in the direction which is transverse to the electron trajectory.