Light induced “Mock Gravity” at the nanoscale

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The origin of long-range attractive interactions has fascinated scientist along centuries. The remarkable Fatio-LeSage’s [1, 2] corpuscular theory, introduced as early as in 1690 and generalized to electromagnetic waves by Lorentz [3], proposed that, due to their mutual shadowing, two absorbing particles in an isotropic radiation field experience an attractive force which follows a gravity-like inverse square distance law. Similar “Mock Gravity” interactions were later introduced by Spitzer [4] and Gamow [5] in the context of Galaxy formation but their actual relevance in Cosmology has never been unambiguously established [6, 7]. Here we predict the existence of Mock-Gravity, “1/r^2”, attractive forces between two identical molecules or nanoparticles in a quasi monochromatic isotropic random light field, whenever the light frequency is tuned to an absorption line such that the real part of the particle’s electric polarizability is zero, i.e. at the so-called Fröhlich resonance [8]. These interactions are scale independent, holding for both near and far-field separation distances.

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The interaction between two objects is usually defined to be long ranged if the force decays with their distance apart, \( r \), as a power law \( \sim 1/r^{n+1} \) with \( n \) smaller than the spatial dimension of the system. Gravity is a typical example of a long-range attractive force in three-dimensions while the interaction between electric or magnetic dipoles (\( n=3 \)) is borderline in between short and long range attraction \([9]\). In contrast, the familiar dispersion forces between non-polar, neutral, molecules and particles, arising from quantum electrodynamic fluctuations, are short range. At close distances the Coulomb interaction between the fluctuating electric dipole moments leads to an interaction energy proportional to \( 1/r^6 \), the so-called van der Waals-London dispersion forces \([10]\). However, when \( r \) is larger than a characteristic resonance wavelength \( \lambda_F \), retardation effects become important since the dipole moments fluctuate many times over the period the light takes to pass between particles. The interaction energy varies then as \( 1/r^7 \) as first shown by Casimir and Polder \([11]\). These interactions can also be derived as a special case of Lifshitz’s theory of attraction between macroscopic bodies \([12]\) in which the force is deduced from equilibrium quantum and thermal electromagnetic field fluctuations \([12–15]\).

In the last years there has been an increasing interest in understanding the non-equilibrium analogs of Casimir forces arising in the interaction between bodies at different temperature \([16, 17]\) like those induced by blackbody radiation from a hot source on atoms and nanoparticles \([18, 19]\). Surprisingly strong long-range interactions between atoms or non-absorbing dielectric particles in a quasi-monochromatic fluctuating random field were predicted \([20–22]\) and experimentally demonstrated for micron-sized particles \([22]\) (similar interactions between pairs of dipoles under the excitation of multiple laser beams were also discussed \([23]\)). Although the effective interaction range can be controlled by the spectral bandwidth of the fluctuating field \([22, 24, 25]\) (with the Casimir-Lifshitz interaction recovered in the limit of a quantum black body spectrum \([22]\)), the existence of three-dimensional artificial gravity like, inverse square law, interaction forces had not yet been demonstrated.

For non-absorbing dipolar particles in a quasi-monochromatic random field, the force always presents a characteristic oscillatory behaviour for distances larger than the light wavelength (reminiscent of a Fabry-Perot-like behaviour). Gravity-like interactions were predicted only for small separation distances \([20, 21, 23]\) assuming that the imaginary part of the polarizability could be neglected, i.e. neglecting radiation pressure effects. However, as discussed below, radiation pressure effects dominate the near-field interactions of non-
absorbing particles, leading to a rather different interaction law. Our main goal here is to show that, in contrast with atoms or dielectric particles, the interaction force between two identical resonant molecules or plasmonic nanoparticles, whose extinction cross section is dominated by absorption, can follow a true attractive inverse square law all the way from near to far-field separation distances. As we will see, the ideal non-oscillating $\sim 1/r^2$ law can only be achieved when the frequency of the random field is tuned to the particles’s Fröhlich resonance (e.g. the Fröhlich frequency, $\omega_F$, of plasmonic silver nanoparticles), clarifying the physical basis of the so-called Mock Gravity and opening the possibility to study (mock) gravitational interactions at the nanoscale. Suspensions of Fröhlich resonant nanoparticles will then offer a promising laboratory for testing the intriguing predictions of the statistical mechanics of systems with long-range interactions [26].

To this end, let us consider two identical nanospheres of radius $a$ separated by a distance $r$ in an otherwise homogeneous medium with refractive index $n_h = 1$. The particles are illuminated by an homogeneous and isotropic random light field consisting of a superposition of unpolarized and angularly uncorrelated plane waves (of frequency $\omega$ and wave number $k = \omega/c$, being $c$ is the vacuum speed of light). If the spheres are sufficiently small, they can be characterized by their electric polarizability $\alpha(\omega)$

$$\alpha(\omega) = \left[ \alpha_0^{-1}(\omega) - i \frac{k^3}{6\pi} \right]^{-1} = |\alpha(\omega)| e^{i\delta_\omega}$$

where $\alpha_0(\omega)$ is a quasistatic polarizability (real in absence of absorption) and $\delta_\omega$ the scattering phase-shift. In order to discuss the “$r$”-dependence of the interaction force given by Eq. (11) in the small particle limit, $ka \ll 1$, we consider $\alpha_0(\omega)$ given by

$$\alpha_0(\omega) = 4\pi a^3 \frac{\epsilon(\omega) - 1}{\epsilon(\omega) + 2}$$

where the particles’ permittivity $\epsilon(\omega)$ is assumed to follow a Lorentz-Drude-like dispersion,

$$\epsilon(\omega) = 1 + \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\omega \Gamma_0}$$

(being $\omega_p$ the plasma frequency, $\omega_0$ the natural frequency and $\Gamma_0$ the damping constant). The polarizability can then be written as [27, 28]

$$\alpha(\omega) = \frac{4\pi a^3 (\omega_F^2 - \omega_0^2)}{\omega_F^2 - \omega^2 - i \left\{ \omega \Gamma_0 + 2(ka)^3(\omega_F^2 - \omega_0^2)/3 \right\}}$$
FIG. 1: **Forces between non-absorbing particles.** Log-log plot of the absolute value of the interaction force for the Lorentz model with parameters $\omega_0 = 0.1\omega_F$, $\Gamma_0 = 0$ and $a = \lambda_F/100$ for (a) the resonant frequency $\omega = \omega_F$ and (b) strongly out of resonance $\omega = 0.1\omega_F$. Red shadowed regions indicate repulsive interaction force.

FIG. 2: log-log plot of the absolute value of the interaction force for the Lorentz model with parameters $\omega_0 = 0.1\omega_F$, $\omega = \omega_F$ and $a = \lambda_F/100$ for different damping coefficients (shown in each plot). Red shadowed regions indicate repulsive interaction force.
where $\omega_F$ is the Fröhlich resonance frequency given by $\omega_F^2 = \omega_p^2 / 3 + \omega_0^2$. The term $-i \Gamma_0 \omega$ accounts for damping by absorption, whereas $-i2(ka)^3(\omega_F^2 - \omega_0^2)/3$ accounts for radiative damping [27].

In absence of absorption, $\sigma_{abs} = 0$, the interaction force (given by Eq. (11a) in Methods), exhibits an oscillatory behaviour in the far-field zone ($kr \gg 1$) with an envelop that decays as $r^{-2}$:

$$\lim_{kr\gg 1} F_{12}(r) \bigg|_{\text{No abs}} \sim -U_E \frac{k^4|\alpha|^2}{2\pi} \frac{\cos(2[kr + \delta_\omega])}{(kr)^2} \frac{r}{r},$$

a result that was first predicted [20] for the interactions between dipolar particles excited by a spatially coherent field after averaging over all orientations of the inter-atomic axis with respect to the incident beam (strictly equivalent to a fixed dimer illuminated by a random fluctuating field [21]). In the near-field zone, $kr \ll 1$, we can distinguish two different regimes. At resonance, $\omega = \omega_F$, the phase shift $\delta_\omega = \pi/2$ and $\alpha(\omega_F) = i6\pi k_F^{-3}$ and the near-field force is repulsive (see Methods), proportional to the energy density of the random field,

$$\lim_{kr\ll 1} F_{12}(r, \omega_F) \bigg|_{\text{No abs}} \sim U_E \frac{k^4|\alpha(\omega_F)|^2}{\pi} \frac{1}{3(kr)^2} \frac{r}{r},$$

being independent on the actual resonant frequency, $\omega_F$, or any other particle’s property. This universal limit had not been noticed previously. In contrast, in the weak scattering limit (strongly off-resonance) when $\omega \ll \omega_F$, as long as $a \ll r$, the interaction force goes as

$$\lim_{ka\ll kr\ll 1} F_{12}(r, \omega) \bigg|_{\text{No abs}} \sim -U_E \frac{k^4|\alpha|^2}{4\pi} \times$$

$$\left\{ \frac{22 \cos(2\delta_\omega)}{15} + \frac{18 \sin(2\delta_\omega)}{(kr)^7} \right\} \frac{r}{r}.$$  

Previous works [20, 21, 23] disregard the last term assuming that, far from resonance, the imaginary part of the polarizability can be neglected (i.e. $\sin 2\delta_\omega \sim 2\delta_\omega \sim 0$ and $\cos 2\delta_\omega \sim 1$) which would lead to an attractive $r^{-2}$, gravity-like, interaction force at short distances. However, even for frequencies strongly off-resonance ($\omega \ll \omega_F$) where $|\alpha| \sim 4\pi a^3$, in absence of absorption Optical Theorem imposes $\sin \delta_\omega = k^3|\alpha|/(6\pi) \sim 2(ka)^3/3$, i.e. $\sin 2\delta_\omega \sim 2\delta_\omega \sim 4(ka)^3/3$. This implies that, for small distances, the attractive term $\sim r^{-7}$ dominates the interaction. These results for non-absorbing particles are summarized in Fig. 1 where we plot
FIG. 3: Real (black line) and imaginary part (red line) of the polarizability versus wavelength in vacuum for a silver nanoparticle with $a = 5\text{nm}$. The interaction forces corresponding to $\lambda = 317, 337, 352$ and $470\text{nm}$ (vertical dashed lines) are shown in Figure 4.

The force (normalized to $F_0 = U_E k^4 |\alpha(\omega)|^2 / (4\pi)$, in our case $F_0 \approx 10^{-18}$) versus separation distance for different illumination frequencies. Forces were calculated from Eq. (11) using the polarizability given by (4). We compare the modulus of the actual force versus distance (in logarithmic scale) with the trends expected for $r^2$ and $r^{-2}$ in Figure 1.a (resonant case) and $r^{-7}$ and $r^{-2}$ in Figure 1.b (out of resonance). Note how a crossover from a $r^{-2}$ to a $r^{-7}$ tendency takes place as the particles get closer. Clearly, except for a narrow window of separation distances, in absence of absorption the interaction forces do not follow an attractive gravity like interaction.

Let us now consider the forces for very small absorbing particles (e.g. few nm sized Ag particles [29]) such that the extinction cross section is dominated by absorption [8], i.e. $\sigma_{\text{abs}} \sim \sigma_{\text{ext}}$. In the weak scattering limit, the interaction force presents again an oscillatory behaviour except at the Fröhlich resonance, where the force can be shown to be given by

$$F_{12}(r, \omega_F) = -U_E \frac{k_F^4 |\alpha(\omega_F)|^2}{2\pi} \frac{1}{(k_F r)^2} \frac{r}{r},$$

i.e. a force that is a non-oscillating long range gravity-like interaction. This equation summarises the most important result of the present work. Notice that within the small particle dipole approximation and for the Lorentz model, the weak scattering limit at the Fröhlich resonance is given by

$$\left(\frac{a}{r}\right)^6 \left(\frac{\omega_F^2 - \omega_0^2}{\Gamma_0 \omega_F}\right)^2 \ll 1.$$
FIG. 4: Forces between Silver nanoparticles. log-log plot of the interaction force for two silver nanoparticles with \( a = 5\text{nm} \) illuminated with an isotropic fluctuating random field of intensity 10W/\( \mu \text{m}^2 \) for (a) \( \lambda = 317\text{nm} \), (b) \( \lambda = 337\text{nm} \), (c) \( \lambda = 352\text{nm} \) and (d) \( \lambda = 470\text{nm} \). Red shadowed regions indicate repulsive interaction force.

and then Equation (9) will hold for distances as small as \( r \sim 3a \) (for shorter distances high order multipoles start being relevant) as long as the quality factor of the resonance \( \frac{\omega_F^2 - \omega_0^2}{\Gamma_0 \omega_F} \) remains smaller than \( \sim 30 \). This is illustrated in Fig. 2, where we show the exact interaction force based on the polarizability given in Eq. (4) for different values of \( \Gamma_0/\omega_F \). Under the condition given by Eq. (10), the interaction turns gravitational-like at all separation distances, all the way from the near to the far field zones.

In order to check the validity of the results in a realistic scenario, we consider the polarizability given by Eqs. (1) and (2) using experimental values for the permittivity of silver nanoparticles [30]. For simplicity, we do not include corrections for nonlocal or size dependent dielectric response. The polarizability of a 5 nm radius silver nanoparticle in vacuum is represented in Fig. 3. Note how the real part of the polarizability is equal to zero at 317
nm and 352 nm. Hence, gravity-like interactions should show up for the two wavelengths at which condition \( \text{Re}(\alpha) = 0 \) is fulfilled. This is indeed what we observe in Fig. 4 where we plot the interaction force given by Eq. (11) for the particular case of silver nanoparticles and compare with the expected behavior given by Eq. (9). Note how for \( \lambda = 317 \) nm and \( \lambda = 352 \) nm the gravitational-like interaction shows up, applying from infinity to short distances until absorption is not large enough to preserve the weak scattering approximation. However, for a wavelength at which the Fröhlich condition is not fulfilled (for instance \( \lambda = 337 \) nm) the gravitational-like behavior disappears yielding to an oscillatory behaviour at long distances. It is worth to emphasise that, for example, gold nanoparticles would not present a clear gravity-like interaction since the real part of the polarizability of a gold nanoparticle does not vanish.

**METHODS**

It can be shown that the averaged force on particle “1” located at \( \mathbf{r} \), due to the presence of particle “2” at the origin of coordinates, can be written as the sum of two terms [21, 22]:

\[
F_{12}(r) = \left\{ \frac{4\pi U_E}{k^2} \right\} \sum_{i=x,y,z} \left[ \text{Im} \left\{ \frac{k^6 \alpha^2 g_i g'_i}{1 - k^6 \alpha^2 g_i^2} \right\} \right] - \left\{ k^2 \sigma_{\text{abs}} \right\} \frac{\text{Re} \left\{ k^3 \alpha \left[ g_i g'_i + g_i g'_i^{*} \right] \right\}}{|1 - (k^3 \alpha)^2 g_i^2|^2} \frac{\mathbf{r}}{r} \tag{11a}
\]

where \( \sigma_{\text{abs}} = k \text{Im}\{\alpha\} - k^4|\alpha|^2/(6\pi) \) is the absorption cross section of a single particle, \( U_E = \epsilon_0 \langle |\mathbf{E}(\mathbf{r}, t)|^2 \rangle /2 \) is the, time-averaged, energy of the fluctuating electric field per unit of volume \( (U_E = U_{EM}/2, \text{being} \ U_{EM} \text{the energy density of the electromagnetic wave}) \), and \( g'_i = \partial g_i / \partial (kr) \) with

\[
g_x(kr) = g_y(kr) = \frac{e^{ikr}}{4\pi kr} \left( 1 + \frac{i}{kr} - \frac{1}{(kr)^2} \right) \tag{12}
\]

\[
g_z(kr) = \frac{e^{ikr}}{4\pi kr} \left( -\frac{2i}{kr} + \frac{2}{(kr)^2} \right). \tag{13}
\]

It is worth to mention that we implicitly assume that the system is in a stationary state and the energy absorbed is transferred to the thermal bath. Since the force is always directed along the radial direction and \( F_{12} = -F_{21} \), the force between two identical absorbing particles in a random, fluctuating field is a *conservative force.*
Equation (11) simplifies considerably in the weak scattering limit, $|(k^3\alpha)g_i|^2 \ll 1$, where recurrent scattering events do not play an relevant role (see the denominators in Eq. (11)). In absence of absorption, and strongly off-resonance ($\omega \ll \omega_F$), the weak scattering approximation holds even at near field distances ($kr \ll 1$), $|(k^3\alpha)g_i|^2 \sim (a/r)^6 \ll 1$, as long as $a \ll r$. However, at the resonance condition in absence of absorption $\alpha(\omega_F) = 6\pi k_F^{-3}$, and recurrent scattering dominate the interaction force in the near field since $|(k^3\alpha)g_i|^2 \sim |6\pi g_i|^2 \gg 1$.

When the extinction cross section is dominated by absorption, $\sigma_{abs} \sim \sigma_{ext}$ the interaction force (11) in the weak scattering limit is simply given by

$$F_{12}(r)_{abs} \sim 4\pi U_E k^4 |\alpha|^2 \sum_{i=x,y,z} \left[ \text{Im} \left\{ \frac{e^{2i\delta_\omega} + 1}{2} g_i g_i' \right\} - \text{Im} \left\{ \frac{e^{2i\delta_\omega} - 1}{2} g_i g_i'^* \right\} \right] \frac{r}{r}.$$  \hspace{1cm} (14)

At resonance $\omega = \omega_F$, $\delta_\omega = \pi/2$ and, taking into account that

$$\sum_{i=x,y,z} \text{Im}(g_i g_i'^*) = \frac{-1}{8\pi^2 (kr)^2},$$ \hspace{1cm} (15)

we obtain the interaction force given in Eq. (9).

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AUTHOR CONTRIBUTIONS

J.L.-H., M.I.M. and J.J.S. conceived the study. J.L.-H. carried out calculations and figures. All authors contributed to the scientific discussion, writing and revising of the manuscript. J.J.S. supervised the study.
COMPETING FINANCIAL INTERESTS

The authors declare no competing financial interests.

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