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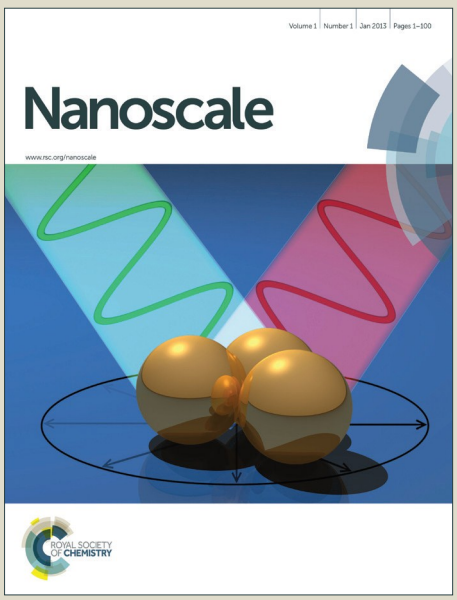

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*Title:***Anderson Localization of light in a colloidal suspension (TiO_2@Silica)**

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Abstract: In recent years, there has been dramatic progress in the photonics field in disordered media, ranging from applications in solar collectors, photocatalyzers, random lasing, and other novel photonic functions, to investigations into fundamental topics, such as light confinement and other phenomena involving photon interactions. This paper reports several experimental evidences of localization transition in a strongly disordered scattering medium composed of a colloidal suspension of core-shell nanoparticles (TiO_2@Silica) in ethanol solution. We demonstrate the crossover from a diffusive transport to a localization transition regime as nanoparticles concentration is increased, and that an enhanced absorption effect arises at localization transition.

Introduction

Scattering media have attracted much attention in recent years, due to potential applications in solar energy, [1, 2]photocatalyzers, [3] random lasers [4], and novel optical devices. [5, 6] The coherent superposition of input fields could give rise to fundamental effects, such as light confinement (localization), [7, 8,9,10,11] enhanced backscattering,[12] much further transmission than the mean free path through the sample, [13] and coherently enhanced absorption.

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[14]Localization of light and a wide variety of associated phenomena have greatly attracted the attention of researchers in the past decades. Localization in a three-dimensional (3D) system could only take place in extremely strongly scattering medium. The requirement is known as the Ioffe–Regel criterion ($kl_s \leq 1$),[15] where $k = 2\pi/\lambda$ and l_s are the wave number and scattering mean free path, respectively. However, critical regime in the approaching to localization (localization transition) has been reported to not so high scattering strengths ($kl_s \leq 5$). [9] In the localization regime, the transmission decreases exponentially instead of linearly with the thickness of a sample (*d*). At the transition, the transmission must have a quadratic dependence on the inverse thickness. [16, 17] This quadratic decay can be interpreted as the onset of localization.However, the residual absorption can provoke a similar decay, which has lead to an important debate.[18,19, 20] In fact, various evidences of Anderson localization have been reported, but questioned. Additionally, according to the theoretical prediction of Sajeev John, an enhanced absorption must arise when the system approaches to mobility edge. [7]Recently, Sperling*et al.* provided evidence for the transition from diffusive light propagation to localization. [10] Significantly, they employed approaches that are intrinsically independent of residual absorption. The localization or localization transition regimes are extremely difficult to be obtained, especially in a colloidal suspension. Particles in a suspension tend to agglomerate and/or precipitate, which causes a decrease in scattering strength. Indeed, to our knowledge, the localization or localization transition regimes have not yet been reported in a colloidal suspension. Ascattering medium in liquid suspension, among others advantages, facilitates the dissolution of molecules, harvesting of products resulting from a photochemical reaction and allows a better study of the Anderson localization itself and associated phenomena. In previous works, [21,22] we introduced ascatteringmedium for a random laser composed of a colloidal suspension of core-shell TiO_2@Silica nanoparticles (Nps), whose silica shell showed an irregular morphology. The silica shell provides a better optical colloidal stability (OCS) and light-coupling enhancement (LCE) with TiO_2 scatter

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cores. Moreover, the silica coatings provide inertness [23,24] and high dispersibility of Nps, [25, 26, 27] which has enabled their use in numerous applications. [28, 29, 30] The effects of OCS and LCE were proposed as likely causes of the increased scattering strength. [21, 22] l_s experimentally determined can be several times lower than $l_s$calculated by Mie-scattering calculation. [22] This fact was explained through a possible light coupling enhancement effect with the TiO_2 cores, due to light refraction at ethanol-silica interface. Here, by using an improved Stöber method, [31, 32] TiO_2nanoparticles were coated with a homogenous silica shell of ~40 nm thickness. A homogeneous silica shell must improve the LCE effect, leading to a significant scattering strength, higher than the reported in our previous works. [21, 22] Here, localization transition was demonstrated by several approaches, among others-measurement of the relative fluctuations of the inverse participation ratio (propagating experiment), inversely proportional to dimensionless conductance, which reflects the extent of localization, even in the presence of absorption. [19] Additionally, an enhanced absorption phenomenon is reported at the localization transition. Localization of light in a colloidal suspension could open new avenues for the designing and manufacture of novel photochemical reactors, powerful sensing tools and others photonic devices based on highly disordered scattering media.

Material and Methods:

Synthesis and characterization of Silica shell:

In the first stage, 5 g of TiO_2Nps were dispersed in 500 ml of spectroscopic ethanol. The suspension of TiO_2 nanoparticles was placed in an ultrasound bath and 6.67 mL of ammonia and 10 mL of TEOS were added. The TEOS and commercial ammonia (NH_4OH 28%-30%) were added alternately in 100 portions of 100 µl and 220 µl, respectively. The synthesized TiO_2@Silica nanoparticle suspension was rota-evaporated and dried in an oven at 70 °C for 2 h. In order to prepare the scattering medium with different [Nps], the TiO_2@Silica powder is re-dispersed in the appropriate ethanol volume. The silica coating on the TiO_2 nanoparticles

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was examined by transmission electron microscopy (TEM) and electron-energy-loss spectroscopy (EELS), using a Carl Zeiss Libra 120 kV transmission electron microscope. The stoichiometric ratio (Ti/Si) of nanoparticles (TiO_2@Silica) was determined by Energy Dispersive X-Ray fluorescence (ED-XRF). The mass percentage ratio (Ti/Si), determined by ED-XRF, was Ti_{72}/Si_{28}. Considering the typical density of the silica obtained by the TEOS hydrolysis around 2 g cm^{-3},[33] we determined an average silica shell thickness of ~42 nm. The sample preparation details for these characterizations and additional TEM images can be found in the supplementary information.

Transport experiments:

For transport experiments, the second harmonic of a *Q*-switched Nd: YAG Continuum Minilite II (25 mJ, 532 nm, pulse width of ~4ns, repetition rate up to 15 Hz, multimode, linearly polarized and spot size of 3 mm) was used and attenuated 10^3 times by neutral density filters (1μJ)(see supplementary information, Figure S2a and S2b, for experimental setups). For transmission coefficient measurements, a CW He-Ne laser, model Uniphase *1125P* (10 mW, 633 nm) was also used. The transmission coefficient (*T(d)*) is defined as the ratio between total transmitted flux and the incident flux.The total transmission is measured with an integrating sphere placed in contact with the back of the sample (fused silica cuvette). For the propagation experiments and coherent backscattering, the same CW He-Ne laser was used (experimental setups, Figure S4 and S5a, supplementary information).

Result and discussion

Silica shell study

Figure 1 shows the TEM image and mapping EELS (Si) of the core-shell TiO_2@Silica NPs. In the TEM image (Figure 1a), the silica coating on the TiO_2 particles shows a regular morphology, with a thickness around 40 nm. Figure 1b shows the Si mapping EELS (orange) of TiO_2@Silica Nps. As it is observed, the silica coating presents great homogeneity. A homogeneous silica shell is expected to improve the LCE effect, leading to increased

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scattering strength. A better definition of the ethanol-silica interface should increase the light refracted toward the TiO_2 scatter core.

Scattering Medium Study

In order to study the scattering medium composed of these core-shell nanoparticles (TiO_2@Silica) in ethanol solution, a set of transport experiments were performed. The transmitted coherent intensity (I_{TC}) and the transmission coefficient (T(d)) were determined as a function of slab thickness (d) for [Nps]of 280, 140, 70, 47, and 14; x10^{10}Nps ml^{-1}. I_{TC} should show an exponential decay (linear in log scale). The l_s values can be determined from slopes. Figure 2a shows the coherent transmission curve (normalized) versus slab thickness for [140x10^{10}Nps ml^{-1}], using the Nd:Yag laser. Three different parts can be distinguished (I, II, and III). In part I, the transmitted intensity decreases more quickly than an exponential. The green line represents the exponential decay ($I_{TC} = e^{-d/l_s}$) determined by fitting with the first experimental points. From the derivative, aninitial ($d$near to zero) scattering mean free path (l_{s0}) is obtained, $l_{s0} = 1.15\mu m$. The red line represents the decay behavior of experimental points. As can be observed, the slope is not constant; it progressively decreases as slab thickness is increased (part I). For slab thickness greater than ~6μm (near point A), the transmitted intensity starts to decay more slowly. This should be because the diffuse intensity (I_{dif}) starts to overweigh the coherent intensity. Near point B, the inelastic scattering should start to play a role, and in part III, the decay is exponential ($e^{-d/l_{MA}}$,blue line).From the inverse slope, we can obtain the macroscopic "absorption" length (l_{MA}), where "absorption" represents the losses by inelastic scattering processes. l_{MA} can be expressed as $l_{MA} = (l_T \times l_{In})^{1/2}$, where$l_T$ and l_{In} are the transport and inelastic mean free path, respectively. $l_T$for d near to zero (l_{T0}), can be determined from the l_{s0} values by $l_{T0} = \frac{l_{S0}}{1-\langle \cos\theta \rangle}$ equation**,** where $\langle \cos\theta \rangle$ isthe average cosine of the scattering angle (see sup. Information). l_{In} near to zero (

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l_{In0}), determined through above equation ($l_{In0} \approx 2680\ \mu m$), is more than three orders larger than $l_{s0} = 1.15\mu m$ and more than 10 times larger than the average photon path length ($l_{eO(Micro)}$) inside the scattering medium before being reflected ($l_{eO(Micro)} \approx 240\ \mu m$). [34]At localization transition,$l_{eO(Micro)}$must be approximately equal or lightly higher than the optical path length of localized photonsinside the scattering medium (microscopic coherence length, ξ_{coh}), since the overall pathlength of non localized photons must vanish when l_T decreases approaching to localization.

Figure 2b shows the coherent transmission curves (part I) for [Nps] of 140, 70, 47 and 14 x10^{10}Nps ml^{-1}. The transmitted intensity decreases more quickly than an exponential for [Nps]≥47x10^{10}Nps ml^{-1}. This effect is pronounced as [Nps] is increased (from 47 to 140; x10^{10}Nps ml^{-1}).This fact represents an anomalous behavior. The green lines represent the exponential decay,determined by fitting with the first experimental points,and red lines represent the experimental behavior. For [280x10^{10} Np ml^{-1}], the intensity in part I decays extremely quickly, which prevented a reliable measurement. For [14 x10^{10} Nps ml^{-1}], I_{TC} decay exponentially. l_{s0} values extracted from the derivative of the green lines are plotted as a function of [Nps] (Figure 2c), revealing that l_{s0} decrease linearly as [Nps] is increased ($l_{s0} \propto 1/[N_{ps}]$).

Part III of the coherent transmission curves are shown in the supporting information (fig. S2 c-g).l_{MA} determined from the inverse slopes (parts III) shows an anomalous behavior as [Nps] is increased. $(l_{MA})^{-1}$ increases more quickly than the expected linear increase (fig.S2h supp. information).Notice that, if $l_{T0}$and l_{In0} would be inversely proportional to [Nps], a $(l_{MA})^{-1} \propto [N_{ps}]$ dependence must be expected.Therefore, in order to study in depth this anomalous "absorption" behavior,l_{In0} values were determinedthrough$l_{MA} = (l_{T0} \times l_{In0})^{1/2}$ equation (table SI supp. information).$(l_{In0})^{-1}$per filling fraction (α_{FF0}) is plotted (fig. 2d left) as a function of the filling fraction ([FF]). For each [FF], α_{FF0} values ("absorption" coefficient) are

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determined from $\alpha_{FF0} = (l_{In0} \times [FF])^{-1}$. In order to determinate l_{In0} for [280x10^{10} Np ml^{-1}], l_{T0} was extrapolated following the $l_{T0} \propto 1/[N_{ps}]$ criterion. Contrary to expectations, α_{FF0} is not constant; it increases quickly in [FF] as a function $\alpha_{FF0} = \alpha_0 + C([FF] - [FF_0])^2$, where C is a constant, and α_0 and [FF_0] are α_{FF0} at the limit of diffusive regime and the critical filling fraction which starts the α_{FF0} anomalous increase, respectively. This anomalous increase of "absorption" coefficient at *d* near to zero represents a very interesting result, which could be strong evidence of the existence of the photon mobility edge. [7, 35, 36] According to the theoretical prediction of Sajeev John, a signature of the electromagnetic mobility edge in a disordered medium is an anomalous rise in energy absorption. [7] Fig.2d (right) shows the enhanced "absorption" factor for *d* near to zero (γ_0) determined by the ratio between effective and classical "absorption" $(\gamma_0 = {}^{\alpha_{FF0}}/_{\alpha_0})$. This γ_0 parameter could be interpreted such that one photon travels an average of γ_0 times around a closed loop path at *d* near to zero, i.e. each photon would interact an average of γ_0 times with the same particles, atoms or molecules within the closed loop paths. This idea could be extrapolated to the elastic polarization of valence electrons to virtual states, which would imply an increase also in the effective refractive index at *d* near to zero (n_{eff0}). This supposed increase of the effective refractive index by localization effect finds a parallel in the dynamic barrier proposed by Campagnano and Nazarov[37] in the border of a disordered electronic medium at localization.

Figure 2e shows the transmission coefficients, measured using two laser sources: one pulsed (Nd:Yag, 532nm) and the other CW (He-Ne, 633nm), for [Nps] of 280, 140, 70, 47 and 14; x10^{10} Nps ml^{-1}. The fluence of the Nd:Yag laser was ~10^7 times higher than of He-Ne laser. For [Nps] ≥ 47x10^{10} Nps ml^{-1}, the transmission coefficient shows a similar quadratic decay $T(d) \propto \beta(d_0 + d)^{-2}$ for the two laser sources, which suggests a quadratic decay seemingly insensitive to wavelength and fluence (in this range). This is exactly the behavior of the scaling theory of localization at the localization transition, predicted by Anderson and

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co-workers. [16, 17] For 14x10^{10}Nps ml^{-1}, the transmission coefficient shows a classical behavior $(d_0 + d)^{-1}$. The green and red solid lines (Figure 2e) are the fitting with experimental points for Nd:Yag and He-Ne lasers, respectively. Notice that for [Nps] ≤ 140x10^{10}Nps ml^{-1}, the transmission coefficients tend to ~(0.47) instead of 1. This effect is explained by the total internal reflection at the output interface air-silica of cuvette (see supporting information for an expanded discussion). For [280x10^{10}Nps ml^{-1}], the transmission coefficient tends to ~0.3 instead of expectedvalue (~0.42), which must be a result of an appreciable increase of the losses due to enhanced "absorption".Notice that, appreciable losses in the light transport must lead to a transmission coefficient lower than expected, especially at d=0. From the quadratic decay of the total transmission,it is difficult to demonstrate being so close to the localization transition, since the losses by the inelastic scattering processes could lead to a similar decay.[18] However, a detailed treatment of the data and an expanded discussion about "absorption" contribution can be found in the supporting information.

From the above results it could be inferred thatan anomalous light transportis reached for [Nps] ≥ 47x10^{10}Nps ml^{-1}.However, l_{T0}values seemtoo high, corresponding to $kl_{T0} > 5$. In order to resolve this mismatch,l_T values were also determined by backscattering method(supp.Information, fig. S5). l_T values, extracted from the angleof backscattering cones for [47, 70, 140 and 280; x10^{10}Nps ml^{-1}], correspond to 0.93, 0.7, 0.64 and 0.47μm, respectively. A simple model for internal reflection was taken into account for correction of l_T values.[38] l_T determined through backscattering method does not decay inversely proportional with [Nps] and these $l_T$values are several times lower than l_{T0} values. This is an anomalous and seemingly contradictory result. Therefore, otherl_T alternativevalues (l_{T*})were also determined considering an increase in the effective refractive index (d near to zero) by localization effect. Let us introduce the following conjecture: if the α_{FF0} increases γ_0 times at d

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near to zero becauseeach photon interacts a average of γ_0 times with the same particles, atoms or molecules (within the closed loop path), then n_{eff0} must also increase as$n_{eff0} = 1 + \gamma_0(n_{eff} - 1)$ (see sup. Information).Thereby, in order to estimate the internal reflection, the effective refractive indexes for each [Nps] were scaled through the enhanced "absorption" factor $\left(n_{eff0} = 1 + \gamma_0(n_{eff} - 1)\right)$ (table SI supp. Information). l_{T*} values for [47, 70, 140 and 280; x10^{10}Nps ml^{-1}], determined through the n_{eff0} correction, correspond to 0.86, 0.6, 0.49 and 0.24μm, respectively. As can be appreciated, bothl_T and l_{T*}, determined by backscattering method (fig.S6, supp. Information), are several times lower than l_{T0}, which represents another interesting result thatcould be explained by anl_T decrease indepth (d).Consequently,the coherent backscattering cone yieldsanl_T value averagedby depth. Of course, the calculus of n_{eff0} and the subsequently determined l_{T*} values maynotbe strictly correct because just at d=0 anadditional increase in the effective refractive indexwould be expected. Notice that,the l_{s0},l_{T0} and $\alpha_{FF0}$are average values determined in a region near to d=0, not at d=0 (interface silica-sample), where these values should increase even more.Additionally, the enhanced factor of coherent backscattering is lower than expected (1.8) for [Nps] ≥ 140x10^{10}Nps ml^{-1}, which could introduce an error in estimating the backscattering cone angle.The $l_T$decrease in depth could bean additional evidence of localization transition.Various authors have theoretically suggested the$l_T$decrease in depth[16,39] at the critical regime of approaching to localization. This fact can be understood by internal reflection of the coherently backscattered photons in the sample input border, which forces the photons path to be longer. Consequently, the likelihood of closed loop path must also be increased. Furthermore, an increase in the closed loop paths must also induce an increase in the effective refractive index at d near to zero (n_{eff0}), which in turn should lead to the internal reflection increasing. The l_T increase for d near to zero would not mean an increase in the physical distance between two successive scattering events; this could be interpretedas an increase in the effective optical pathlength between two

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successive scatterings, because the effective refractive index must increase for *d* near to zero.Of course this effect would be appreciable when the coherently backscattered light (closed loop paths) starts to have an appreciable contribution.Obviously, the influence of internal reflection decreases with depth; however, it should increase as angle of incidence is increased.The above effects can be understood such that the density of localized states must increase in the vicinity of the boundary, which was theoretically predicted by Mirlinin disordered electronic media. [39, 40]Other perspective about this issue was provided by Ramos and co-workers, [41] who demonstrated that the presence of a finite barrier in the input border provokes an increase of the quantum interference (localization) in a disordered electronic medium. The slope decrease with depth in the coherent transmission curves (part I) for [Nps] $\geq 47 \times 10^{10}$ Np ml^{-1} could be a consequenceofl_s decrease in depth. The last has an important physical connotation because it would mean that for large*d,*the effective "absorption" coefficient(α_{FF}) could be even lower than the classical "absorption" coefficient(α_0).This would imply that the photon cloud propagation does not fill completely all the sample volume, which means that for large*d*, the photons are propagated by specific paths,i.e. like a much further transmission than the mean free path through the sample [13]and/or a subwavelength light localization. [42]This means that, only those photons with very specific entry phase (φ) into the scattering medium (position (x,y,*d*=0)) could reach large *d*, and that at position (x,y,*d*) such photons would be strongly correlated. However, at different positions (x,y,d)-(x´,y´,d) in the *d* plane the photons would be completely uncorrelated (localization). Therefore, for a largeenough *d*, the photon paths would cease to be equally probable. This statistical problem at the mobility edge in a 3D system was addressed for the first time by Altshuler et al.,[43] who proposed a Poisson lawbecause the paths would be completely uncorrelated.

Propagating experiment

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In order to corroborate localization transition, another one experiment has been performed. The intensity structure of a probe beam (He-Ne laser) was measured after propagating a distance *d*=1.8mmthrough the scattering medium. A CCD camera collected the image of intensity profile $I(x,y)$ at the sample output face (figure S4 supp. information). The input probe beam is <100 μm full-width at half-maximum (FWHM)and is perpendicular to the sample surface. For the five[Nps], the normalized intensity profiles at samples output are displayed in figure 3. As can be observed, the beam intensity structure narrows as the [Nps] is increased. For 14x10^{10} Nps ml^{-1}, a Gaussian beam profile $e^{-2(r/\sigma)^2}$ is observed, where *r* is the radial distance of the beam center. However, for [Nps] ≥ 47x10^{10} Nps ml^{-1}, the beam profile ceases to be Gaussian, it could be fitted with a $e^{-2\left(|r|/\sigma\right)^{1+\nu}}$ function, where $0 < \nu < 1$. For each [FF], the confinement of the beam at the output plane is quantified by the inverse participation ratio $P \equiv [\int I(x,y)^2\, dxdy]/[\int I(x,y)\, dxdy]^2 = \frac{1}{\pi}[\int I(r)^2\, dr]/[\int I(r)\, dr]^2$, which has units of inverse area, and an effective width $\omega_{\text{eff}} = (P)^{-1/2}$. If absorption is negligible, σ in the fitting function $e^{-2\left(|r|/\sigma\right)^{1+\nu}}$ must be proportional to ω_{eff}($\sigma \propto \omega_{\text{eff}}$). Figure3f shows the effective width at the samples output and ν value (extracted from fitting), as a function of filling fraction, revealing that ω_{eff} and ν decrease monotonically as the filling fraction is increased.Notice thatfor [280x10^{10} Nps ml^{-1}] (fig.3e), theintensity profileshows a rounding at *r* near to zero, and for larger*,*it decreases more quickly than the $e^{-2\left(|r|/\sigma\right)^{1+\nu}}$ fittingfunction (red solid line).This effect can be associated to an increase in the losses by enhanced "absorption". In this fitting, ν was fixed and its value was taken by extrapolation (~0.28). σvalue was also fixed following the same ($\sigma \propto \omega_{\text{eff}}$) criterion. Figure 3g shows the corresponding value of the inverse participation ratio (P)as a function of the filling fraction, along with its statistical standard deviation (marked by error bars). The

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statistical standard deviation (ΔP) is determined by $\left(\frac{1}{\pi}\sqrt{\left(\frac{d[P(r)]}{dr}\right)^2} \times unitdistance\right)$evaluated in $r \approx 0$. Figure 3h presents the relative fluctuations of the inverse participation ratio, $\Delta P/P$, as a function of filling fraction. As can be observed, $\Delta P/P$ value increases with the filling fraction.For a filling fraction of 3.5 %, equivalent to 47x10^{10}Np ml^{-1}, $\Delta P/P = 0.67 \cong 2/3$, which corresponds to the onset of localization suggested by Genackand co-workers. [19]The $\Delta P/P$ increase agrees with the prediction [16,39]that the relative fluctuations in *P* are inversely proportional to the dimensionless diffusion coefficient, which must tend to unity ("absorption" negligible) when the system approaches to localization (Poisson law).For [280x10^{10} Nps ml^{-1}], $\Delta P/P$ is lightly above unity, which must be a consequence of "absorption". [19]The above result representsan additional evidence of localization transition. However, the"absorption"couldalso have influence over the beam propagation. But, there is not aneasy analytical expression forthe dependence of"absorption" with angle at the localization transition regime. One can think that the "absorbed"light would increase with *r* because the effective photon pathlength increases with *r*. However, the "absorbed"lightcould increase even more with *r* at localization transition because the enhanced "absorption" factor can increase with the incidence angle.Notice that, the incidence angle should influence over the photons transportat localization transition because the internal reflection must increase as incidence angle is increased regarding to normal incidence. This fact might increase the likelihood of closed loop paths, leading to a conductance decrease.Therefore, an additional experiment was performed in order to confirm localization transition.The probe beam was introduced in the scattering medium at 30º, 45º and 60º regarding to normal incidence. The intensity profiles $I(x, y)$ at the sample output face were collected for [Nps] of: 14 and 140; x10^{10} Nps ml^{-1}.If ω_{eff} decrease in [Nps]would be caused by the "absorption", ω_{eff} must be

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independent toincident anglefor the two[Nps].Instead, if ω_{eff}decrease is basically a consequenceof the approaching to localization, the profilewidth shoulddecrease as incidence angle is increased, i.e. a narrowing of the intensity profile must be observed.Notice that an increase in the incidence angle must provoke an increase in the internal reflection of the coherently backscattered photons, i.e. an increase in the effective barrier at the input border. Consequently, an increase in localization must be expected. [41]This would mean a decrease of diffusion coefficient with incidence angle. This effect would be strong evidence of the approaching to localization, since incidence angle should not be an influence on the "absorption" or in the light transport at diffusive regime. Figures 4a-bshowa comparison of the normalized intensity profiles for [14 and 140; $x10^{10}$ Nps ml^{-1}] and incidence angles of 0º and 60º. As can be clearly observedfor [$14x10^{10}$Np ml^{-1}](diffusive regime), the incidence angle has noinfluenceon the intensity profile.However, a narrowingof the intensity profile is observed for [$140x10^{10}$Np ml^{-1}](localization transition) as incidence angle is increased, revealing alinear ω_{eff}decrease (fig. 4c).This isstrong evidence of an anomalous transport.Note thatfor the incidence angle of 60º and [$140x10^{10}$ Nps ml^{-1}] (fig.4b), the intensity profile is sharpened at r near to zero. This could be caused by a decrease of diffusion coefficient. Additionally, the intensity profile decreases more quickly than the fitting function $e^{-2\left(|r|/\sigma\right)^{1+\nu}}$ at large *r*(red solid line). Thelast could be a consequence of the increase of enhanced"absorption" factor (γ_0) with incidence angle, which must be caused by an increase in the likelihood of closed loop paths by the internal reflection. On the other hand, the narrowing of the intensity profile,asangle of incidence is increased, would be further evidence that the "absorption" effect is negligible for [Nps] ≤$140x10^{10}$Np ml^{-1}; such as was inferred from the propagation experiments and total transmission. An expanded study of propagationand "absorption" as a function of incidence angle and [Nps] is called for, in order to understand in depth the photons transport at localization and the associated phenomena to

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it.From the above results, a clear transition is observed for [Nps] ≥ 47 x10^{10}Nps ml^{-1}, which corresponds with a $kl_{T*} \approx 8.6$. This critical value is higher than that reported by Sperling et al. ($kl_T \approx 4.5$). [10]Note that for [47 x10^{10}Nps ml^{-1}], the filling fraction is only 3.5%. In turn, the critical regime is reached for a filling fraction ~10 times lower than that reported by Sperlinget. al. The higher filling fractionin their samples combined with the high powers used in their experiments must lead to an increase in the inelastic scattering processes. [11, 44]Probably for this reason, they did not observe an appreciable change in the statistical distribution of the photon cloud. The losses by inelastic scattering must provoke a smoothingin the maximum of intensity profile, appearing like a Gaussian distribution.Additionally, a high filling fraction combined with very high power should also give rise a significant increase in the nonlinear refractive index, leading to an increasing of the photon-photon interaction. [45]The latter must lead to a decrease in the coherence length, ξ_{coh}. Consequently, a higher scattering strength would be required in order to get the critical regimen of localization transition.

Conclusion

Core-shell TiO_2@Silica nanoparticles with an appropriate silica shell (thickness and homogeneity) allowed us to obtain a liquid suspension with a significantly high scattering strength. We report several evidences of the critical regime of localization transition in a colloidal suspension, which is achieved at [Nps] ≥ 47 x10^{10}Nps ml^{-1}. For [Nps] ≥ 47x10^{10}Nps ml^{-1}: The transmitted ballistic intensity starts to decay more quickly than an exponential, the "absorption" coefficient (near to zero) shows an quadratic increase in [Nps](instead a constant value),from which an increase of refractive index also close to zero is proposed; the transmission coefficient starts to show a quadratic decay and a Gaussian probe beam that is propagated through the samples showed a $e^{-2\left(|r|/\sigma\right)^{1+\nu}}$ $(0 < \nu < 1)$ intensity profile at sample output, instead a Gaussian profile. We also must highlight that for a probe beam inclined

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regarding to normal incidence, the intensity profiles are similar for [$14x10^{10}$Np ml^{-1}](diffusive regime). However, for [$140x10^{10}$Np ml^{-1}], a narrowing of the intensity profile is observed as incidence angle is increased. Additionally, an l_T decrease in depth for [Nps] $\geq$ 47 $x10^{10}$Nps ml^{-1} is inferred from the measurements of coherent backscattering and ballistic transmission (part I).

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Author Contributions

All authors conceived, designed, performed and analyzed the experiments; E.J-V, P.C.O, W.M.F. and G.F. de S. contributed materials/analysis tools; E.J-V guided the research and wrote the manuscript.

Additional Information

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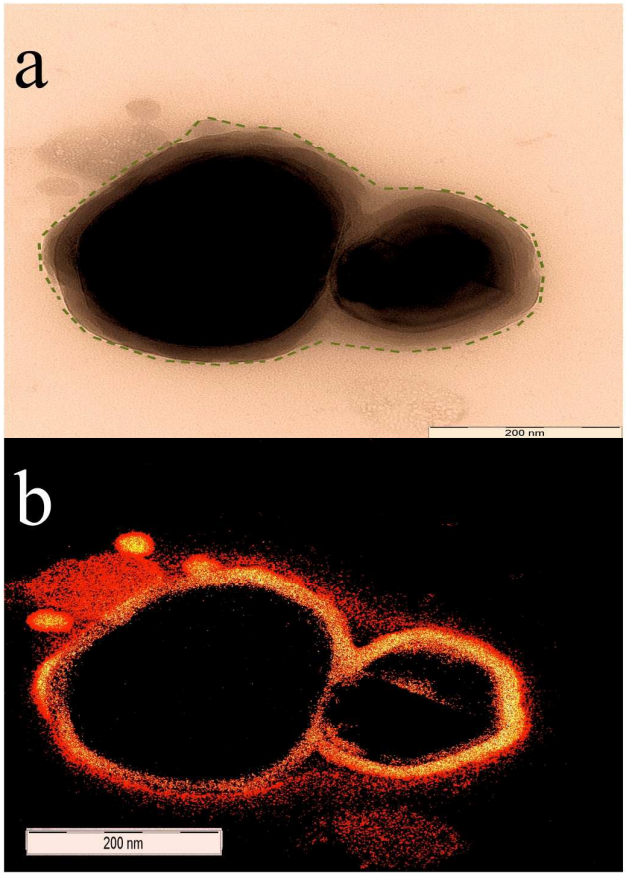

Figure 1.Characterization of TiO_2@Silica nanoparticles via transmission electron microscope.a) TEM image and b) EELS-TEM (Si) chemical mapping of core-shell TiO_2@Silica Nps demonstrates the presence of silicon (orange) onto the TiO_2 surface. The dotted green line (a) indicates the outer surface of the silica coating. The scale bar corresponds to 200 nm.

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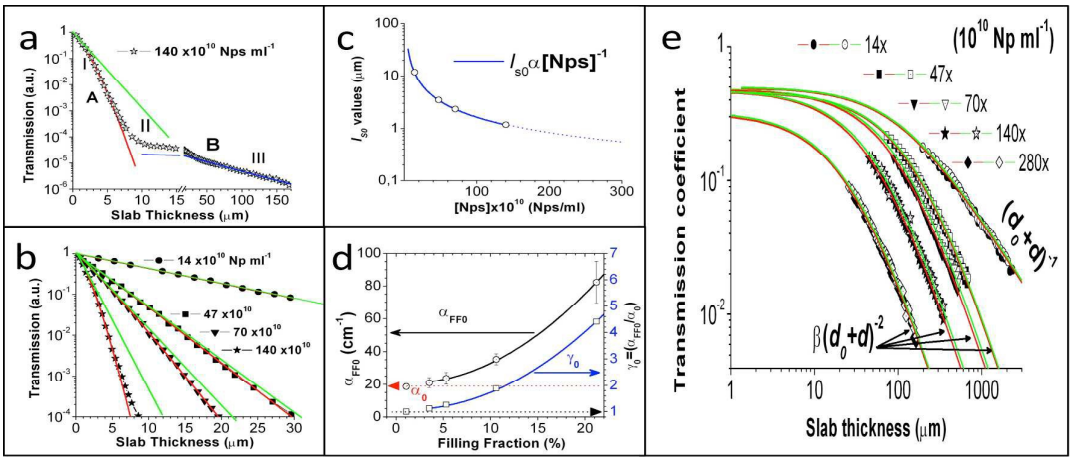

Figure 2. Transport experiments. a)Transmitted coherent intensity (I_{TC}) versus slab thickness [140x10^{10}Nps/ml] (blue line, fitting at part III), and b) I_{TC} for [Nps]: 14, 47, 70, and 140; x10^{10}Nps ml^{-1}. The red and green lines represent the decay behavior of experimental points and the exponential fitting with the first experimental points, respectively. c) l_{s0} versus [Nps], d) anomalous increase of: α_{FF0} (left) and the γ_0 (right) as [FF] is increased, red dotted line represents α_0 value, error bars correspond to fitting errors. e) Transmission coefficient for the lasers: pulsed Nd:Yag and CW He-Ne and [Nps] of: 14, 47, 70, 140 and 280; x10^{10}Nps ml^{-1}. Open and filled symbols correspond to experimental points for Nd:Yag and He-Ne (lasers), respectively. The green and red solid lines represent the fitting with experimental pointsfor Nd:Yag and He-Ne (lasers), respectively.

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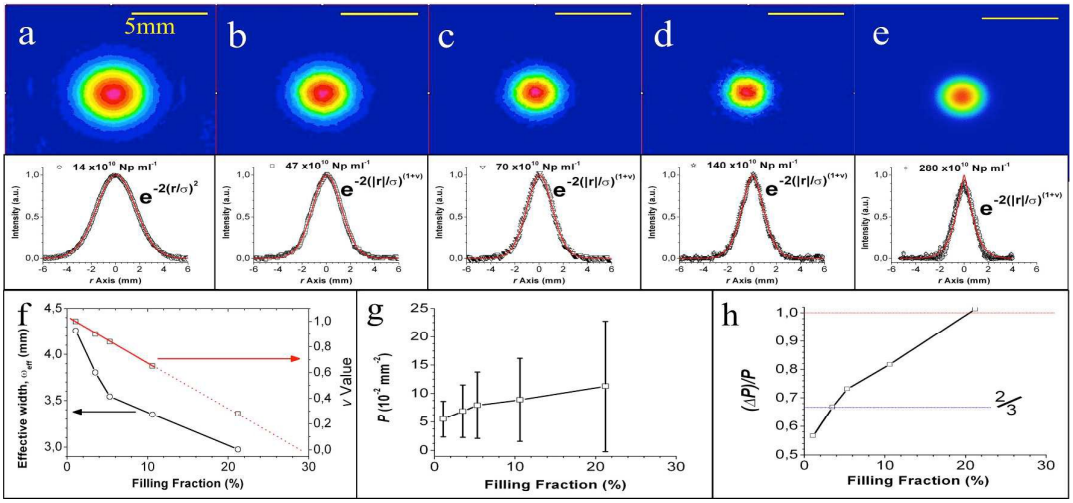

Figure 3.Experimentally measured intensity distributions at the samples output for [Nps]: a) 14, b) 47, c) 70, d) 140and e) 280; x10^{10}Nps ml^{-1}. The propagation length of probe beam was 1.8 mm and scale bar represents 5mm. For [14x10^{10}Np ml^{-1}], the intensity profile fits very well with a gaussian profile $\boldsymbol{e^{-2(r/\sigma)^2}}$. In b), c), d) and e) ($[\mathbf{Nps}] \geq \mathbf{47x10^{10}\ Np\ ml^{-1}}$), the intensity profiles could be fitted to $\boldsymbol{e^{-2\left(|r|/\sigma\right)^{1+\nu}}}$, where $\mathbf{0 < \nu < 1}$. f) ω_{eff} (left) and ν (right, red) versus [FF]. g) Inverse participation ratio *P* as function of [FF].The error bars are the statistical standard deviations of *P* $\left(\Delta \boldsymbol{P} = \langle \frac{\mathbf{1}}{\boldsymbol{\pi}}^{\mathbf{2}} \sqrt{\left(\frac{\partial P(r)}{\partial r}\right)^{\mathbf{2}}}\ (\boldsymbol{unit\ distance}) \rangle\right)$ around $\boldsymbol{r = 0}$. h) Relative fluctuations of the inverse participation ratio, $\left(\Delta \boldsymbol{P}/\boldsymbol{P}\right)$, as a function of [FF]. The increase monotonically of $\left(\Delta \boldsymbol{P}/\boldsymbol{P}\right) > \mathbf{2}/\mathbf{3}$, and the transition from the gaussian curve of a) to the $\boldsymbol{e^{-2\left(|r|/\sigma\right)^{1+\nu}}}$ decaying curves of b), c), d) and e) display the crossover from diffusive transport a) to the localization transition regime.

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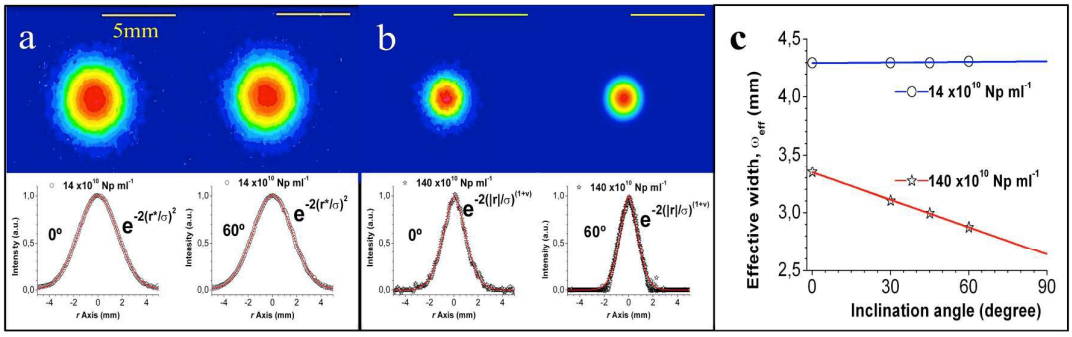

Figure 4.Propagating experiment as a function of incidence angle for: a) [14x10^{10}Np ml^{-1}], the output intensity profileis independent to incidence angle, equal Gaussian profiles$e^{-2(r/\sigma)^2}$ are observed; b) [140x10^{10}Np ml^{-1}], the intensity profile narrows as incidence angle is increased, showing a decrease in the photon conductance. c) ω_{eff}monotonicallydecreases with incidence angle for [140x10^{10}Np ml^{-1}], showing that photon transport is determined by localization phenomenon, not by "absorption", as already was previously inferred.

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