

Localization of light: a power sensing tool

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Abstract: Localization of light induced by correlation in the scatterers' positions is achieved in an ordered photonic colloidal (TiO₂@Silica) suspension. A strongly enhanced light-matter coupling is observed as the concentration of TiO₂@Silica is increased above threshold.

Introduction: Anderson localization of light in three–dimensions (3D) (true Anderson localization) shows prospects for entirely new optical phenomena [1,2], which might lead to the generation of new-type photonics devices. The tendency towards localization at correlated long-range hopping in higher dimensional systems, demonstrated theoretically by Kravtsov and co-workers and called “*correlation-induced localization*”, has revealed the universality of this phenomenon [3]. Historically, various pioneering experiments that studied the transmission of electromagnetic waves through strongly scattering optical media have claimed the observation of Anderson localization of light [4–6]. However, these works were questioned firstly by opponents [7,8] and later refuted by their authors [9,10]. Inelastic scattering processes (absorption or nonlinearity) lead to a decrease in the photon coherence length, hampering the interference effects required for localization [11,12], making direct observation of localization difficult and elusive. In fact, according to the theoretical prediction of Sajeev John [13] and our experimental results [14–17], an enhanced absorption arises when the system approaches localization. The strategy used in these previous works was to increase the scattering strength by increasing the scatterers' concentration (randomly distributed scatterers) in order to reach the Ioffe-Regel criterion for localization ($kl_T \leq 1$), where $k=2\pi/\lambda$ and l_T are the wave number and transport mean free path, respectively. However, the increase of scatterer concentration (decreasing the scatterers separation) favors the near field coupling, which can hamper localization of light [18]. Another alternative strategy to reach localization should be achieved by introducing a correlation in the scatterers positions, that is, inducing certain order in the disorder or, in other words, to introduce disorder in an ordered photonics structure (photonic crystal), which was suggested early by Sajeev John [19]. Recently, Dal Negro and co-workers have predicted theoretically that aperiodic correlations used for the engineering of photonic media (e.g. metallic nanostructures) are suitable photonic platforms for localization of light, revealing strongly enhanced light-matter coupling compared to the traditional periodic and homogeneous random media [20,21]. During the last five years, we have been studying the localization of light (3D) in a colloidal system composed by core-shell TiO₂@Silica nano particles (NPs) suspended in ethanol [14–17,22–24]. In a previous work [17], we proposed that the correlation in the scatterers' position due to the long-range interaction (Coulomb) might favor significantly photon interference (localization), which would explain the anomalous transport of light observed in our core–shell TiO₂@Silica suspension in ethanol, even for $3 < kl_T < 8$. The correlation in the scatterer's positions induced by a long-range Coulomb interaction between scatterers (TiO₂@Silica NPs) is due to the electrostatic field (ζ -potential) provided by the silica shell [23,24]. According to the colloidal-classical theory of Derjaguin-Laudau-Verwey-Overbeck (DLVO) [25,26], the pair-wise particle interaction arises from the interplay of attractive van der Waals forces (F_{attr}) and repulsive Coulomb forces (F_{rep} , double layer force) screened by the Debye-Hückel ions' cloud. The modulo of both potentials increases when the separation between particles is decreased and/or the Debye length (λ_{Debye}) and ξ -potential increases. In this research, we study the light-matter coupling by Raman scattering in this colloidal system composed by TiO₂@Silica (Rutile@Silica) NPs suspended in ethanol. The hydrodynamic diameter ($D_{Hyd}=890$ nm), proportional to λ_{Debye} , and the ξ -potential (-70 mV) of TiO₂@Silica NPs suspended in ethanol, which gives an estimative of the extension range of the Coulomb interaction, were determined by dynamic light scattering (DLS) at very low [TiO₂@Silica].

Results: A Micro-Raman XploRA Horiba with a CW 532 nm laser as an excitation source was used for Raman scattering measurements. A detailed description of the experimental setup can be found in ref. [23]. The Raman scattering signal of the TiO₂@Silica NPs suspensions were collected for TiO₂ filling fractions (FF_{TiO_2}) from 0.26% up to 12.1%. Two strong peaks located at around 445 and 610 cm⁻¹ assigned to the E_g and A_{1g} modes of rutile are observed (Figure 1a). The intensity of the Raman peaks (Figure 1b) increases quicker than linearly as FF_{TiO_2} is increased above a specific value (FF_{onset}), revealing an enhancement of the Raman signal per particle (Figure 1c). This anomalous enhancement of light-matter coupling above FF_{onset} can be explained by the localization phenomenon and, interpreted as localized photons interacting several times with the same particles, molecules or atoms within the

localized states [14], which is a consequence of the strong photon correlation [1,2]. This phenomenon was predicted theoretically by Sajeev John [13] and demonstrated experimentally by us [14,16,17,24].

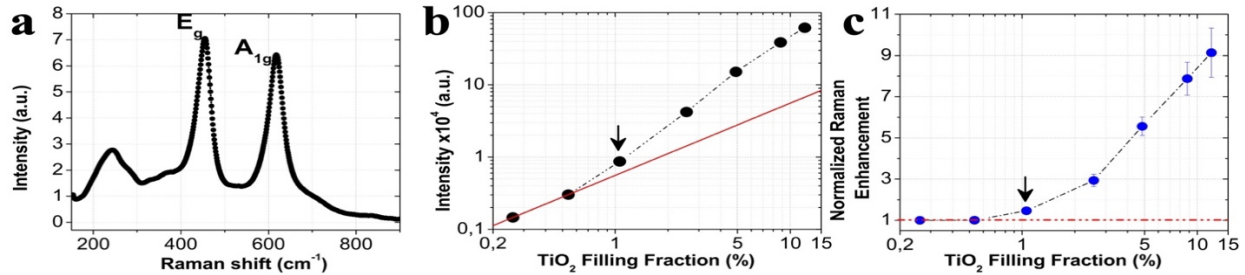


Figure 1. a) Raman spectrum collected from the surface of the $\text{TiO}_2@$ Silica suspension (rutile peaks; E_g , A_{1g}). b) Intensity of the Raman peak (E_g ; 445 cm^{-1}) as a function of FF_{TiO_2} increases more quicker than the expected linear behavior (represented by red line). c) Intensity of Raman peak normalized by FF_{TiO_2} shows an enhancement above $FF=1.06\%$ (represented by black arrow).

Conclusion

Strongly enhanced light-matter coupling is shown by means of the strong enhancement of the Raman signal per particle, revealing a new optical phenomenon that opens up new possibilities for the designing and manufacture of powerful sensing tools.

References

- S. John, "Localization of Light," *Phys. Today* **44**(5), 32 (1991).
- S. John, "Why trap light?," *Nat. Mater.* **11**(12), 997–999 (2012).
- P. A. Nosov, I. M. Khaymovich, and V. E. Kravtsov, "Correlation-induced localization," *Phys. Rev. B* **99**(10), 104203 (2019).
- D. S. Wiersma, P. Bartolini, A. Lagendijk, and R. Righini, "Localization of light in a disordered medium," *Nature* **390**(6661), 671–673 (1997).
- M. Störzer, P. Gross, C. M. Aegerter, and G. Maret, "Observation of the critical regime near anderson localization of light," *Phys. Rev. Lett.* **96**(6), 1–4 (2006).
- T. Sperling, W. Bühner, C. M. Aegerter, and G. Maret, "Direct determination of the transition to localization of light in three dimensions," *Nat. Photonics* **7**(1), 48–52 (2013).
- F. Scheffold, R. Lenke, R. Tweer, and G. Maret, "Localization or classical diffusion of light?," *Nature* **398**(6724), 206–207 (1999).
- F. Scheffold and D. Wiersma, "Inelastic scattering puts in question recent claims of Anderson localization of light," *Nat. Photonics* **7**(12), 934 (2013).
- T. Van Der Beek, P. Barthelemy, P. M. Johnson, D. S. Wiersma, and A. Lagendijk, "Light transport through disordered layers of dense gallium arsenide submicron particles," *Phys. Rev. B - Condens. Matter Mater. Phys.* **85**(11), 1–11 (2012).
- T. Sperling, L. Schertel, M. Ackermann, G. J. Aubry, C. M. Aegerter, and G. Maret, "Can 3D light localization be reached in "white paint"?", *New J. Phys.* **18**(1), 13039 (2016).
- A. A. Chabanov, M. Stoytchev, and A. Z. Genack, "Statistical signatures of photon localization," *Nature* **404**(6780), 850–853 (2000).
- F. Evers and A. D. Mirlin, "Anderson transitions," *Rev. Mod. Phys.* **80**(4), 1355–1417 (2008).
- S. John, "Electromagnetic absorption in a disordered medium near a photon mobility edge," *Phys. Rev. Lett.* **53**(22), 2169–2172 (1984).
- E. Jimenez-Villar, I. F. da Silva, V. Mestre, P. C. de Oliveira, W. M. Faustino, and G. F. de Sá, "Anderson localization of light in a colloidal suspension ($\text{TiO}_2@$ silica)," *Nanoscale* **8**(21), 10938–10946 (2016).
- E. Jimenez-Villar, V. Mestre, W. S. Martins, G. F. Basso, I. F. Da Silva, and G. F. De Sá, "Core-shell $\text{TiO}_2@$ Silica nanoparticles for light confinement," *Mater. Today Proc.* **4**(11), 11570–11579 (2017).
- E. Jimenez Villar, M. C. S. Xavier, J. G. G. S. Ramos, N. U. Wetter, V. Mestre, W. S. Martins, G. F. Basso, V. A. Ermakov, F. C. Marques, and G. F. de Sá, "Localization of light: beginning of a new optics," in *Complex Light and Optical Forces XII*, D. L. Andrews, E. J. Galvez, and J. Glückstad, eds. (Proceeding SPIE 10549, 2018), p. 1054905.
- E. Jimenez-Villar, M. C. S. Xavier, N. U. Wetter, V. Mestre, W. S. Martins, G. F. Basso, V. A. Ermakov, F. C. Marques, and G. F. de Sá, "Anomalous transport of light at the phase transition to localization: strong dependence with incident angle," *Photonics Res.* **6**(10), 929 (2018).
- S. E. Skipetrov and I. M. Sokolov, "Absence of Anderson Localization of Light in a Random Ensemble of Point Scatterers," *Phys. Rev. Lett.* **112**(2), 023905 (2014).
- S. John, "Strong localization of photons in certain disordered dielectric superlattices," *Phys. Rev. Lett.* **58**(23), 2486–2489 (1987).
- R. Wang, M. Röntgen, C. V. Morfonios, F. A. Pinheiro, P. Schmelcher, and L. D. Negro, "Edge modes of scattering chains with aperiodic order," *Opt. Lett.* **43**(9), 1986–1989 (2018).
- F. Sgrignuoli, R. Wang, F. A. Pinheiro, and L. Dal Negro, "Localization of scattering resonances in aperiodic Vogel spirals," *Phys. Rev. B* **99**(10), 104202 (2019).
- E. Jiménez-Villar, I. F. da Silva, V. Mestre, N. U. Wetter, C. Lopez, P. C. de Oliveira, W. M. Faustino, and G. F. de Sá, "Random Lasing at Localization Transition in a Colloidal Suspension ($\text{TiO}_2@$ Silica)," *ACS Omega* **2**(6), 2415–2421 (2017).
- V. A. Ermakov, W. S. Martins, N. U. Wetter, F. C. Marques, and E. Jiménez-Villar, "Localization of light induced in ordered colloidal suspensions: powerful sensing tools," *Nanoscale* **13**(13), 6417–6425 (2021).
- C. T. Dominguez, A. A. V. Gomes, N. U. Wetter, J. Dipold, V. Mestre, W. S. Martins, and E. Jiménez-Villar, "Random lasing at localization induced in correlated colloidal system," *Opt. Mater. (Amst.)* **120**, 111428 (2021).
- R. J. Hunter, *Foundations of Colloid Science*, 2nd ED. (Oxford University press, 2001).
- O. L. Sánchez-Muñoz, J. Salgado, J. Martínez-Pastor, and E. Jiménez-Villar, "Synthesis and Physical Stability of Novel Au-Ag@SiO₂ Alloy Nanoparticles," *Nanosci. Nanotechnol.* **2**(1), 1–7 (2012).