Non-linear optical effects in colloidal suspensions in the continuous wave regime

A simple setup for undergraduate laboratory experiments

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Research context and introduction

Introduction and motivation

- Development of a low-cost experimental setup to highlight the optical non-linear effects of nanoparticle solutions
- Extension of the work carried out by *Turchiello et al* [1] on the non-linear optical (NLO) properties of the soy sauce in the continuous wave (*cw*) regime
- Use of gold nanoparticles (GNPs) as a thermal lens
- Study of the role of the synthesis media on the NLO properties of GNPs

GNPs preparation

- **GNPs synthesis method:** Two-steps method using (1) Spherical GNPs as seed and (2) surface catalyzed reduction of Au^{3+} by NH_2OH [2]
- **Synthesis parameters:** Amount of added 0.025M $HAuCl_4$, presence or not of NH_2OH, presence or not of poly(vinyl) alcohol (PVA, 6% w:w) in the synthesis solution.

Nanoparticles synthesis scheme

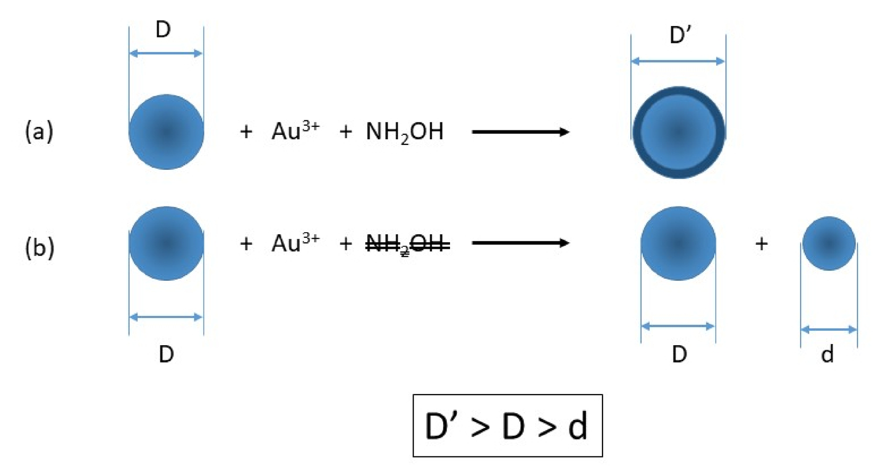

Figure 1: Synthesis scheme for the nanoparticles (a) with NH_2OH (b) without NH_2OH (redrawn from Brown (1998)).

Experimental setup for NLO characterization

- **Principles of the Z-scan method [5]** On the optical axis, the sample translates through the focus of the lens. During the translation, the power transmitted is measured either entirely (*open aperture*) or partially, through an aperture (*closed aperture*), by photodiodes. Open and closed aperture modes highlight two- or multi-photon absorption and nonlinear refraction effects, respectively.

- **Beam waist:**

$$W(z) = w_0 \left(1 + \left(\frac{z}{z_0}\right)^2\right)^{1/2}$$

with z_0: Rayleygh length. At the focus $W_0 = 72.6 \pm 1.4\ \mu m$ and at the sensor location $W(z_{sensor}) = 1805 \pm 46\ \mu m$.

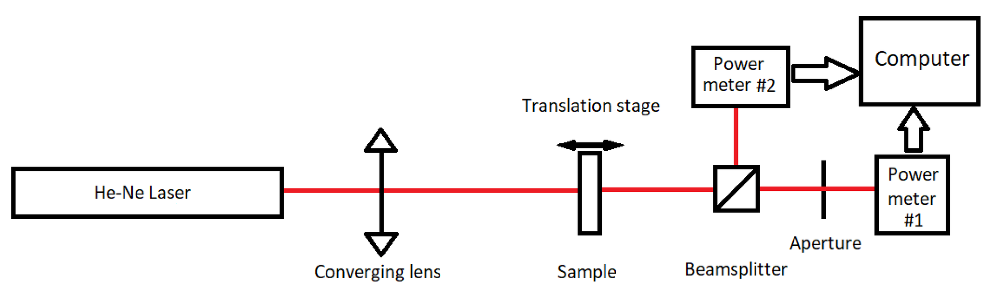

Figure 2: Z-scan experimental setup. At first the He-Ne laser beam is focused by a lens. After its passage trough the sample, the beam is divided by a beamsplitter and collected by two power meters. $\lambda_{He-Ne} = 632.8\ nm$, $P_{He-Ne} = 12.7\ mW$ and $f_{lens} = 10\ cm$

Results and discussion

Linear optical properties

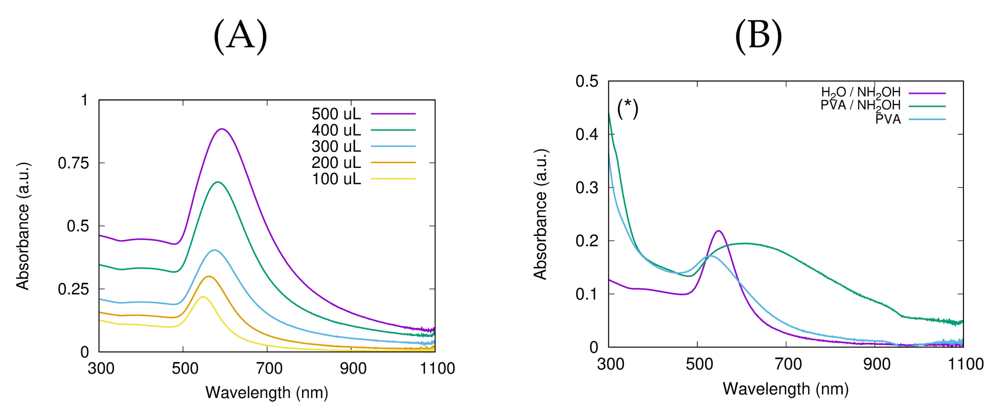

Figure 3: Linear optical properties of the GNPs. (A) Effect of the amount of added $HAuCl_4$ (0.025M) on the absorbance in the presence of NH_2OH (B) Effect of PVA and of NH_2OH on the absorption spectrum

- Increasing the added amount of $HAuCl_4$ increases the absorbance and the scattering of the colloidal solutions (Fig. 3A)
- Red shift of the peak corresponds to larger GNPs
- Adding PVA (6% w:w) slows down the reduction (remaining reagents : * in Fig. 3B)
- NH_2OH effect combined to PVA: important enlargement of the size distribution (Fig.3B, green curve)

Added $HAuCl_4$	100 uL	200 uL	300 uL	400 uL	500 uL
λ_{SPR} (nm)	548	561	575	583	593
Abs_{max}	0.219	0.300	0.404	0.674	0.885
Width (nm)	107	133	171	190	222

Table 1: Resonance parameters changes as a function of the amount of added 0.025M $HAuCl_4$ (Fig. 3A)

Nonlinear optical properties

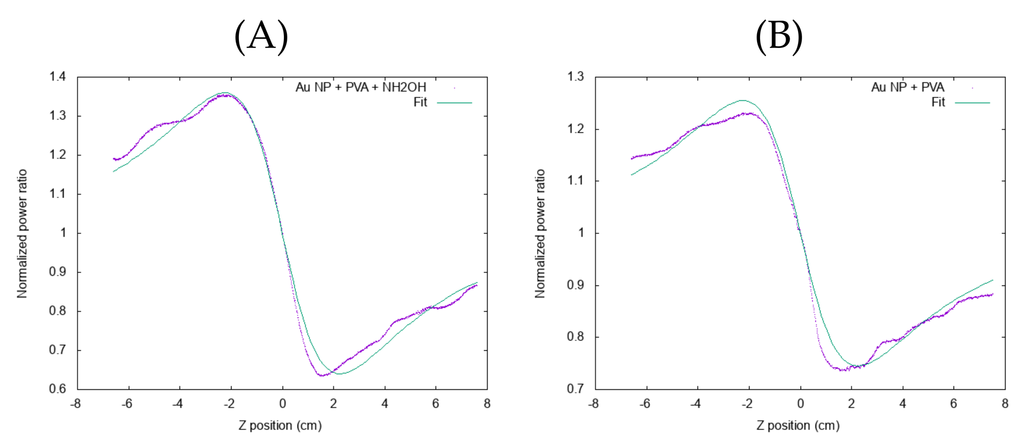

Figure 4: Z-scan of GNPs (A) with NH_2OH (B) without NH_2OH

- Ratio between transmittance in closed and open aperture to remove nonlinear absorption contribution
- Fit formula:

$$T(z) = 1 + \frac{4\frac{z}{z_0}\Delta\phi_0}{\left[1 + (\frac{z}{z_0})^2\right]\left[9 + (\frac{z}{z_0})^2\right]}$$

- Peak-to-valley signals seen from Fig. 4A and B correspond to negative $\Delta\Phi_0$
- Self-defocusing nature of the gold nanoparticles ($n_2 < 0$)

NP's	with NH_2OH	without NH_2OH
$\alpha\ (mm^{-1})$	0.729	0.564
$n_2\ (m^2/W)$	$-3.55 \cdot 10^{-10}$	$-1.43 \cdot 10^{-7}$

Table 2: Absorption coefficient and nonlinear refraction index

Far-field images

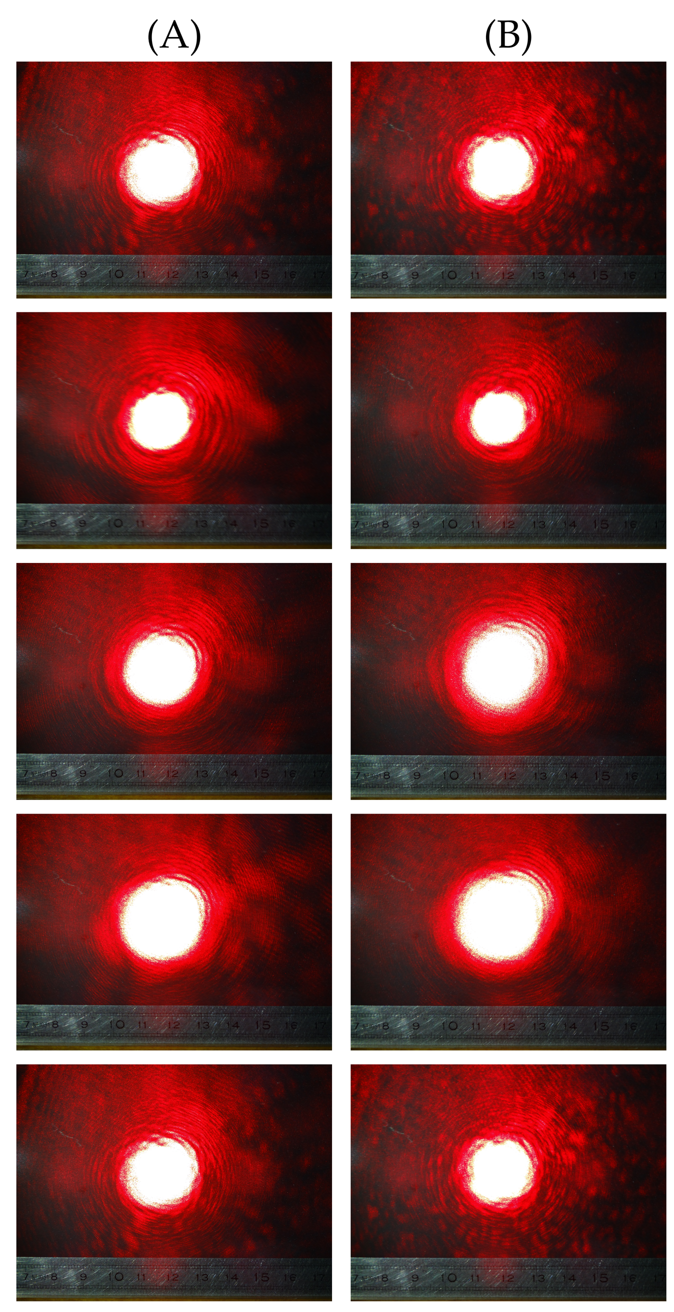

Figure 5: Far-field images of the beam at different sample positions (Distance from the focus: 2.4 m, Image width: 10 cm). From top to bottom: z = -6.6 cm, Peak, Focus, Valley, z = 7.6 cm. (A) GNPs without NH_2OH (B) GNPs with NH_2OH

Conclusions

- **Nonlinear optical effects of gold nanoparticles produced with a low-cost setup were shown in transmission and are visible in the far-field images**
- **Proof that the gold nanoparticles are self-defocusing; even with the add of PVA and NH_2OH which created gold nanorods**
- **Thermal lens property of the gold nanoparticles were highlighted**

References

[1] R. de F. Turchiello, L.A.A. Pereira, and S.L. Gómez, *Am. J. Phys.*, 85 (2017) 522.

[2] K.R. Brown, and M.J. Natan, *Langmuir*, 14 (1998) 726.

[3] R.W. Boyd, Nonlinear Optics, Academic Press, 2008.

[4] R.A. Ganeev, Nonlinear Optical Properties of Materials, Springer, 2013.

[5] M. Sheik-bahae, A.A. Said, and E.W. Van Stryland, *Optics Letters*, 14 (1989) 955.

Acknowledgments

JC, GS and LW equally contributed to this work.