Holographic study of heterogeneous dynamics by Amplitude Time Resolved Correlation (ATRC)

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Abstract: ATRC is a new ligth scattering technique that measures by holography the field amplitude scattered by a sample, and analyze its spatial correlations. ATRC outperforms other light scattering techniques like DLS, DWS and TRC.

OCIS codes: 090.1995: Digital holography; 290.5820: Scattering measurements; 030.6140: Speckle; 110.6150: Speckle imaging; 100.4550: Correlators; 040.2840 Heterodyne

Citation

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Dynamic light scattering (DLS) [1], diffusing wave spectroscopy (DWS) [2] and time resolved correlation (TRC) [3] are well-established techniques for investigating the dynamics of a wide variety of systems in physics, chemistry, biology, and medicine. DLS and DWS consider the intensity time fluctuations of the scattered light, and study samples, whose dynamics are homogeneous in time. TRC considers the spatial fluctuations, and is able to study slow or temporally heterogeneous dynamics. We propose, in this work, to replace the TRC multi pixel detector by an holographic one, and to consider the spatial fluctuations of the field complex amplitude E as a new quantity of interest. We introduce thus a new tool called ATRC for Amplitude Time Resolved Correlation, which outperforms TRC and open new perspectives.

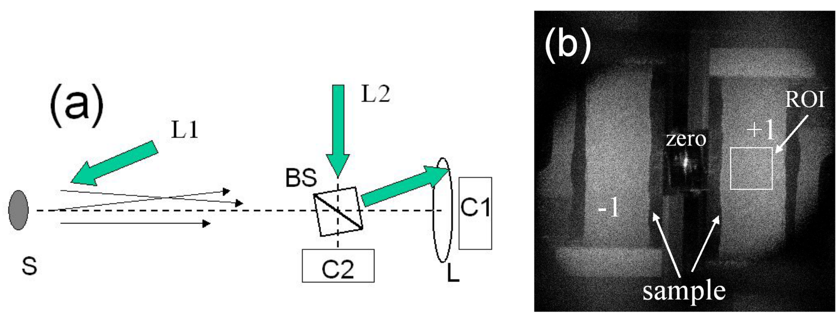

Fig. 1. (a) TRCA typical setup. (b) Reconstructed image of a nanocomposite sample.

Fig.1 (a) shows a simplified diagram of the TRCA setup. The sample S is illuminated by the laser beam L1. The light scattered by the sample is split by the beam splitter BS into two imaging paths, corresponding to cameras C1 and C2. Lens L made the image of the sample S on C1, which records the light intensity image I of S. On the other hand, the camera C2 records the interference pattern (i.e. the hologram) of the scattered light with the reference laser light L2, and the complex amplitude image E of S is calculated by holographic reconstruction. The optimize the detection sensitivity [4], holography is heterodyne [5] and off-axis .

Fig.1 (b) shows the intensity reconstructed image ($I = |E|^2$) of a 30×10 mm^2 Styrene-Butadiene/Silica nanocomposite sample, which is observed in reflection. Because 2 phase detection is used, both the +1 and -1 image of the sample are seen (right and left hand side images of Fig.1 (b)). To analyze the dynamic of the sample, TRC considers

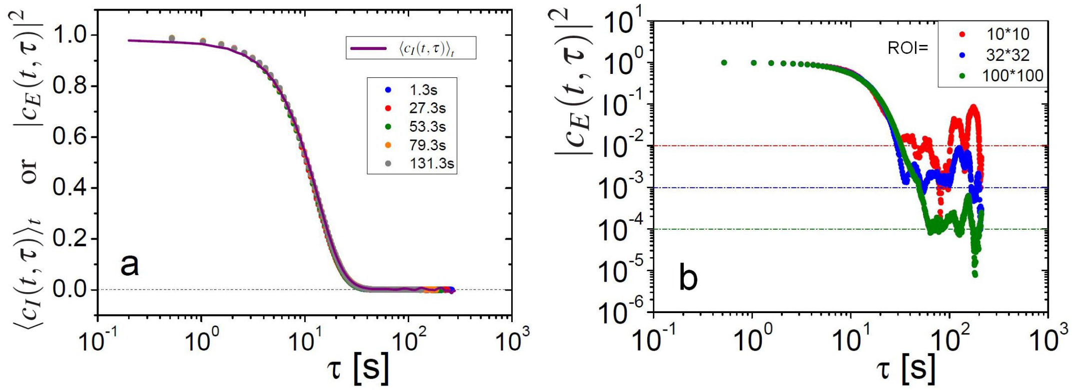

Fig. 2. (a) Log-lin plot of $\langle c_I(t,\tau)\rangle_t$ measured by TRC (purple solid line), and $|c_E(t,\tau)|^2$ measured by ATRC for $t = 1.3, 27.3, 53.3, 79.3$ and 131.3 s (full circles). (b) Log-log plot of $|c_E(t,\tau)|^2$ measured by ATRC. The different curves correspond to different size of ROIs. Dash-dotted lines represent the $1/N_{\mathrm{ROI}}$ expected background noise level.

the 2 times intensity correlation within a region of interest (ROI):

$$c_I(t,\tau) = \frac{\left\langle I_p(t) I_p(t+\tau)\right\rangle_p}{\left\langle I_p(t)\right\rangle_p \left\langle I_p(t+\tau)\right\rangle_p} - 1\,. \tag{1}$$

where $I_p(t)$ is the intensity of the p-th pixel at time t, and $\langle\cdots\rangle_p$ indicates an average over the ROI pixels. For a stationary sample, $\langle c_I(t,\tau)\rangle_t = g_2(\tau) - 1$, where $\langle ...\rangle_t$ is the time average, and $g_2(\tau)$ is the 1 time intensity correlation function considered in DLS and DWS. Since holography allows to measure the field E within the ROI, we have introduced the 2 times field correlation:

$$c_E(t,\tau) = \frac{\left\langle \tilde{E}_p(t)\tilde{E}_p^*(t+\tau)\right\rangle_p}{\left\langle|\tilde{E}_p(t)|\right\rangle_p \left\langle|\tilde{E}_p(t+\tau)|\right\rangle_p} \tag{2}$$

which is related to the 1 time field correlation function $g_1(\tau)$ by $\left\langle|c_E(t,\tau)|^2\right\rangle_t = |g_1(\tau)|^2$. Although $|g_1|^2 = g_2 - 1$ in the ideal case, measuring c_I or $|c_E|^2$ is not equivalent. Indeed, for a fully developed speckle, and for an average over N_{ROI} pixels, the measurement noise floor is $\sim\sqrt{1/N_{\mathrm{ROI}}}$ for c_I, and $\sim 1/N_{\mathrm{ROI}}$ for $|c_E|^2$. It results that the sensitivity is much better when using the complex amplitude E measured by holography.

ATRC has been experimentally validated with a colloidal suspension made of 1.14 μm diameter melamine particles (Microparticles GmbH) dispersed in a water-EmkaroxTM mixture (15/85 wt%), whose scattering properties are invariant with time. Figures 2 (a) shows $\langle c_I(t,\tau)\rangle_t = g_2(\tau) - 1$ measured by TRC with camera C1, and $|c_E(t,\tau)|^2$ measured by ATRC with camera C2 at fixed time $t = 1.3, 27.3...131.3$ s. As seen in Fig. 2 (b), the TRC and ATRC measurements are equivalent, but ATRC is less noisy, since the ATRC results are obtained without averaging over time t. Figures 1 (d) plots $|c_E(t,\tau)|^2$ in log-log scales, for $N_{\mathrm{ROI}} = 10^2$, 32^2 and 10^4. As expected, the background noise scales like $1/N_{\mathrm{ROI}}$.

ATRC proposed here outperforms other light scattering techniques like TRC and open new perspectives in the study of slow and heterogeneous dynamics.

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References

1. B. Berne and R. Pecora, "Dynamic light scattering," Wiley, New York, USA (1976).
2. D. Pine, D. Weitz, P. Chaikin, and E. Herbolzheimer, "Diffusing wave spectroscopy," Physical review letters **60**, 1134 (1988).

3. L. Cipelletti, H. Bissig, V. Trappe, P. Ballesta, and S. Mazoyer, “Time-resolved correlation: a new tool for studying temporally heterogeneous dynamics,” Journal of Physics: Condensed Matter **15**, S257 (2002).
4. M. Gross and M. Atlan, “Digital holography with ultimate sensitivity,” Optics letters **32**, 909–911 (2007).
5. F. Le Clerc, L. Collot, and M. Gross, “Numerical heterodyne holography with two-dimensional photodetector arrays,” Optics letters **25**, 716–718 (2000).