Image-time Correlation and Its Applications

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ABSTRACT

In this paper, Image-time correlation is a beneficial technique to apply various applications of dynamical events, where temporal changes of positions of particles (or objects) can be quantified, constructed by the collection of time-resolved video data. With the fast development of camera technologies and imaging softwares, the optimization of time-binning and spatial-resolution are much friendly-used for the imaging analysis. Time-dependent different regions of interest (ROIs) can be chosen, such that the time resolution of subsequent images should be at least 10 times faster than the actual dynamical events. Then time-correlation function will be calculated by the 2d intensity-intensity correlation of an each pixel-pixel of individual time frame of image, and the initial slope of correlation function gives a characteristic time scale that measures the average magnitude of particle velocities within the field-of-view. The advantage of image-time correlation is easy to implement as compared to the PIV (particle imaging velocimetry). In this short proceeding, the performance of image-time correlation is highlighted through a number of applications; such as the slowing down behaviors of collective dynamics of orientation textures, dimer-oscillations in rod-networks, and clay (platelet) migrations in an oil-water droplet. In each system, the interpretation of image-time correlation is addressed, based on the experimental observations.

CCS Concepts

• Information systems → Computing platforms

Keywords

Image-time correlation; reconstruction of time-lapsed image; decay rates; field-induced criticality; collective dynamics; dimer-oscillations in rod-networks; clay migrations in an oil-water droplet.

1. INTRODUCTION

Image-time correlation is originally developed by the needs of experimental analysis on low frequency electric field-induced dynamical states of charged fibrous virus-suspensions, where the various stable phase transitions are observed in the suspensions of fd virus at a low ionic strength [1]. Fig. 1 is the electric-field induced phase/state diagram of charged fibrous virus suspensions,

in the field amplitude and frequency plane. The suspensions of fd-virus were prepared and purified following standard biological protocols; using the XL1 blue strain of *E. coli* as the host bacteria [2]. The length of fd-virus is 880 nm with a core diameter of 6.7 nm, and the persistence length is about 2200 nm, covered with highly negatively charged fd-coat proteins [3, 4]. In particular, dynamical states that are found, above certain field amplitude, as a sharp first-order like transition, where small nematic domains melt and reform, kinetically (20s-2s). The dynamics of melting and formation of N-domains are accelerated on increasing the field amplitude just above the N_D^* -to-Ds transition line. The image-time correlation is used successfully to predict the field-induced criticality of the charged fd-virus suspensions [5], where much statistical processes are needed to estimate for such complicated phenomena. The characteristic time constant is then a measure for time with which N-domains melt and form, and the decay time constant is probed as a function of the field amplitude, especially at the N_D^* -to-Ds phase transition, as well as the Ds-to- D_f transition line. The characteristic time diverges on approaching the N_D^* -to-Ds phase transition on lowering the field amplitude. However, the transition from the Ds-to- D_f -state is defined as the point where the sharp decrease of time constant with increasing field amplitude ceases to occur, and the dynamics becomes essentially independent of the field amplitude.

Figure 1. Electric-field induced phase/state diagram of charged fibrous virus suspensions, in the field amplitude and frequency plane: N-phase: coexistence of an isotropic and non-chiral nematic phase, N^* -phase: chiral-nematic phase, N_D^* -phase: disconnected chiral-nematic phase, Ds- and D_f -state: a slow- and fast-dynamical state where non-chiral domains melt and reform, H-phase: a homogeneous phase, where the fd-rods are aligned along the electric field.

2. PRINCIPLE OF IMAGE-TIME CORRELATION FUNCTION

To construct the intensity-time traces of images, the intensity matrix values of every pixel AOI (1 pixel $\times$ 1 pixel) are reconstructed for dynamic image correlation function, as a

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“readable” Ascii format to correlate with different time frames of AOI. The modified intensity matrix is defined as the subtracted intensity average value of AOI pixel for every time frame from the raw intensity data. Fig. 2 shows the simple sketch of the enhanced contrast between “bright” and “dark” intensity AOI pixels. The modified intensity matrix is then reconstructed as a building block of individual time frame, such that the each time frame image is subtracted with the overall average intensity value of the initial time frame. Then enhanced contrast is sketched as a bright (white) and dark (or black) unit pixel area, against the mean average of a gray color code. In principle, for a long time of duration, the change of the dynamical events can be completely “independent”, so that at far later time after the event occurs, the time-correlation of dynamical states is expected to be “non-correlated” at the end of time event. This is briefly depicted in the long time later after an event occurs, which has now shown flipped from “white” to “black” and “black” to “white” code. The right panel of Fig. 2 is shown for the correlation function of typical intensity-traces for both slow and fast dynamical state, up and down figures, respectively. The apparent intensity oscillations are due to the spatio-temporal motion of dynamical states of the small nematic domains (of fd-virus particles). The correlation function is defined as in Fig. 2 right; where I is the transmitted intensity at a given pixel and the brackets $\langle \dots \rangle$ denote averaging over all pixels. We made a program to calculate the dynamical image correlation function. We have found the best performance of the intensity averaging is 1 pixel by 1 pixel, as compared to 5 pixels by 5 pixels (or 10 pixels by 10 pixels) averaging. This may be related to a low magnification objective lens (10X objective) that is chosen for larger sampling of AOI in our system shows the typical correlations functions of our dynamic imaging correlations as a function of increasing field amplitude (as indicated the arrow direction). Here, the relaxation time of dynamical states of melting and forming of small nematic-domains of fd-virus particles is presented as a function of applied field amplitude at a low frequency (of 10 Hz). It turned out that this time binning may not be fast enough to characterize “completely” in the fast dynamical states, where the relaxation behaviors could be realized at much microscopic time scale. As a consequence, the background value turned out to be occasionally be- low zero, indicating that the time step is too large to carry on the local events correlations. It would be desirable when the time binning of the sampling time has to be tested with the background value to present as the dynamical state. Also it has to be chosen as much faster exposure CCD camera to ensure the exposure time of camera setting is much faster than the actual duration time for measurement. Thus by allowing the time step is much smaller than the change of visual morphological changes in dynamical states, time-lapsed images are recorded within the chosen frame setting, and saved (or exported) as time-sequential bmp files with 8 bit gray data that is mostly common format in handling image objects.

3. APPLICATIONS OF THE IMAGE-TIME CORRELATION

3.1 Field-induced dynamical states of small nematic domains (~30 μm size)

Field-induced phases in the phase diagram of Fig. 1 are stable, due to interactions of charged fd-viruses, by changing the applied field amplitude and frequency. Thus kinetics of field-induced dynamical states are interested in the quantification with image-time correlations: The simple scheme of an image-time correlation

is drawn in Fig. 2, with its definition, $C_I(t)$, where $I(t)$ is the intensity of a pixel at time t , while the brackets $\langle \dots \rangle$ indicate averaging over all pixels. A typical correlation function is sketched briefly on the right side of Fig. 2, as it decays from the value of 1 to 0 at later time, when the system is statistically equivalent. The image-time correlation function can be then fitted with the stretched exponential function, shown in Fig. 3, where B , A , τ , and β are fitting parameters for the background, amplitude, decay time, and the stretching exponent power, respectively. Here the variations in intensity traces within the D_F -state are much faster as compared to the D_S -state in Fig. 1. Therefore, time traces of the transmitted intensity through crossed polarizers, as detected by the CCD camera for a single pixel, are collected to calculate the image-time correlation functions. In the proposed algorithm, in Fig. 2, each time frame of image is then reconstructed with its own intensity average at a fixed time frame, before the time-correlation is taken place throughout the whole time-lapsed image frame numbers. The correlation function carries out nicely on the decay rates of any dynamical events, including the polydispersity of evolving systems. Fig. 3 is shown for characteristic features of image-time correlations for different applications, discussed in following subsections: (a) slow dynamics of orientation textures of charged fd-viruses, (b) polymerized polystyrene dimer-oscillations embedded in biphasic fd-virus networks, and (c) local migration of clays in oil-water droplet interface, comparing het hydrophilic versus hydrophobic clays.

Figure 2. Simple scheme of the definition for an image-time correlation function of $C_I(t)$, and t_w is waiting time window.

3.2 Slow dynamics of collective orientation textures: Evolution of decay rates

Recently, a lyotropic glass transition is observed in the crowded suspensions of charged fd-viruses at a low ionic strength (of 0.16 mM Tris/HCl buffer) [6]. Right after filling the sample to the optical cuvette, shear-induced orientation textures extend over very large regions with a spatial extent of the order of hundreds of microns that are induced by shear alignment (see Fig. 3(a)). For the lower fd-concentrations (of 3.8 mg/ml), these large nematic domains break up into smaller domains after a few hours. In contrast, for higher concentrations (of 12.3 and 13.6 mg/ml), the initially shear-induced large domains do not break up into smaller domains, even for longer waiting times of up to about 50–100 hours. In order to quantify the slow dynamics of this orientation texture, image-time correlation functions are performed, in a long-time series of images, typically taken every 10–15 minutes, over a time span of about two weeks. The transmitted intensity is recorded and read by a single pixel on the CCD-camera chip at time. The correlation function depends on both the waiting time,

which is the time after filling the sample, and the “delay time”, which is the characteristic relaxation time of the correlation function, is measured at that waiting time, which is now depicted in Fig. 3(a). In these collective slow dynamics of orientation textures, only the initial decay rate is valid through as the inverse initial slope of image time-correlation functions. Then the total delay-time span of a measurement is about to be 30–50 hours. Furthermore, for the lower concentrations, the decay rate is large for short waiting times and decreases with increasing waiting time to reach a constant value after 80–100 hours, determined as the equilibrium time by image-time correlations.

However, for the larger fd-concentrations, there are no waiting-time dependences of the correlation function, right from the start after filling the cuvette. Thus, a qualitatively different behavior for the larger concentration of 13.6 mg/ml is observed and detected in image-time correlations: The decay rate is now a constant, independent of the waiting time. The texture is now essentially frozen at this larger concentration. Moreover, the values for decay rates are now much smaller when compared to the equilibration-plateau values for the two lower concentrations. There is a quite sharp change of the behavior of the texture dynamics as the concentration is increased from 11.1 to 12.3 mg/ml. Thus the concentration, where the texture freezes is thus found to be accurately (as 11.7 $\pm$ 0.6 mg/ml) [6, 7]. The small, but still a finite value of the decay rate above the texture-freezing concentration is due to the slow release of local stresses within the initially shear-aligned regions. Qualitative correlations are sketched for evolving in time is indicated as Fig. 3(a). Therefore, with image-time correlations, the orientation texture dynamics are distinguished by either the system is reached to the equilibrium (in below the glass concentration) or the “freezing” state (of the glass-concentration).

3.3 Dimer-oscillations in an isotropic-nematic biphasic-rod network

Brownian motions are commonly observed in any thermodynamic systems, and so do for the functionalized polystyrene (PS) spheres in the monophasic (of isotropic- or nematic-phase). However, interestingly, in isotropic-nematic (fd-virus) biphasic rod-networks, synchronization of PS dimer-oscillation occurs [8]. This observation is useful to provide some aspect of effective electrostatic interactions of charged complex system of spheres in the host of I-N biphasic rod networks: PS microspheres are prepared by dispersion polymerization of purified styrene monomer, with polyvinylpyrrolidone (molecular weight $M_w \sim 55$ Kg/mol), 2-azobisisobutyronitrile, and ethanol used as a stabilizer, initiator, and dispersion medium, respectively [9]. The images of Fig. 3(b) show typical repetition of synchronized oscillations, with the morphology of negatively charged PS spheres (with a diameter of 1.5 μ m) dispersed in an isotropic-nematic coexistent rod-networks at a low ionic strength (0.032 mM) Tris-HCl buffer. The system is shown to spontaneously form dimers, which exhibit a synchronized oscillatory motion. The synchronization of oscillations occurs in bulk states, in the absence of an external field. The synchronization of PS-dimer oscillation is driven by a delicate balance between the short-attractive “twisted” interaction of PS-dimers and long ranged electrostatic repulsive interactions of charged fd-rods. The interest is then the local enhancement of “twist-nematic” elasticity in reorienting of the dimer oscillations. The analysis of image-time correlations are provided with the data movies and Fourier transforms of averaged orientations for the synchronized oscillations of dimers in biphasic I-N coexistence concentration of charged fd-virus suspensions [8].

The quantification of kinetics of dimer oscillations are then performed by an image-time correlation function: When the electric-field is turned on, rather slower decay rate of different steps of dimer oscillation is appeared, however as soon as a low electric field is applied, a fast decay occurs immediately and progresses further to a lower value for different states of synchronized dimer oscillations. The image-time correlation functions are increased again in which gives certainly non-zero background value, shown in Fig. 3(b), which means that the PS-dimer oscillations in a biphasic fd-rod network are fundamentally determined by the stability of a coexistence concentration, set by a thermodynamic equilibrium [8]. The synchronization is occurred by the collective motions of I-N biphasic phase, long-ranged repulsive electrostatic interactions in the isotropic-nematic biphasic network of charged fd-viruses and the short ranged attraction of PS-dimers. This finding is then useful to design novel microscopic motors in demixing complex fluids.

Figure 3. Characteristic performance of image-time correlation functions for different applications of the system: (a) slow dynamics of orientation textures of charged fd-viruses, (b) polymerized polystyrene dimer-oscillations embedded in biphasic fd-virus networks, and (c) local migration of clays in oil-water droplet interface, where the upper is the hydrophilic (MMT) and lower is the hydrophobic (OMT) clays, and the sedimentation direction is towards the right.

3.3 Local migration of clays in oil-water droplet: hydrophobic versus hydrophilic clays

Another application of the feasibility of an image-time correlation is tested to probe the local interfacial migration velocities of clay particles in the oil-water droplet interface. Here, two different types of clays are used; one for hydrophilic (MMT) and the other is hydrophobic (OMT), where the characterization of local interfacial migration of clay particles within an oil droplet in an aqueous environment is extracted, shown in Fig. 3(c). Details of oil-water intracranial tensions are important topics for the stabilization process in food industry research. Pickering emulsions prevent coalescence of droplets [10, 11], typically, emulsions are fabricated by turbulent mixing for a short time, which are subsequently stable for long times in the quiescent state. The kinetic pathway to prepare Pickering emulsions is crucial for their long-term stability [12].

Local migrations with different Region Of Interest (ROI), after the formation of an oil droplet, can be determined by both the initial slope and the baseline of image-time correlation functions, for the local migrations of clays, as well as their constant value at large time values for an indicator of the concentration of mobile clay particles. Thus, a higher value of the baseline of correlation functions is interpreted as relatively build-up of immobile clays, i.e., a lower concentration of mobile clay particles. Most intriguing finding is the influence of the Marangoni effect on the migration velocities of the hydrophilic (MMT) clay particles, shown in the upper images of Fig. 3 (c), which adsorb at the oil-water interface, contrary to the nonadsorbing hydrophobic (OMT) clays (in lower of Fig. 3(c)). The diminishing particle concentration for the MMT clays is attributed to adsorption, by which particles disappear from the ROIs. The amplitudes for the OMT clays remain constant, as the increase of the concentration due to sedimentation is relatively small during the measuring time. Furthermore, the interfacial migration of the MMT clays increases with increasing waiting time up to 200 s, for different ROIs, which is again in contrast to what is seen for the OMT clays. The increasing average migrations for the MMT clays are attributed to the Marangoni effect, where the motion of particles within the interface drags particles in the vicinity of the interface along through the induced flow. The Marangoni-induced migrations are apparently much stronger than typical sedimentation velocities, which determine the initial values to the decay rates before adsorption occurred. Then the interfacial migration due to Marangoni forces of clay particles within an oil droplet, immersed in water, is relevant for the formation kinetics of Pickering emulsions. The initial decay rate and the baseline of these correlation functions depend on the waiting time in qualitatively different ways, in Fig. 3 (c). Throughout the analysis of image-time correlation, the estimated sedimentation velocities of both MMT and OMT clays are indeed quite small; the Marangoni-induced back flow for the hydrophilic (MMT) clays is much pronounced than that of the hydrophobic (OMT) clays. A quasi-stationary state is reached for waiting times larger than approximately 200s. Flow that is induced by rising particles within the interface due to Marangoni forces cause particles at the sides of the droplet to attain a relatively large velocity as compared to their sedimentation velocity.

4. CONCLUSIONS

In this short paper, three different applications of the image-time correlations are introduced with their performances to characterize the variance of dynamics, either the fast- or the slow-dynamics, represented in Fig. 3. The above examples and interpretations of image-time correlation for the slowing down behavior of collective dynamics, dimer-oscillations in rod-networks, and clay (platelet) migrations are not only applicable to describe their own dynamics, but also to bring “predictive” kinetics of the dynamical events. Furthermore, field-induced dynamics would be certainly valuable subjects to be explored in broader interest of other experimental realizations to compare the model-based theories.

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