

Evidence of pattern formation in the Langmuir Reverse Schaefer films of liquid crystal incubated in silver nanocolloids: A combined experimental and theoretical study

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Spontaneous pattern formation at unstable fluid-fluid interfaces is a popular feature of nonequilibrium systems, often arising from the interplay between interfacial tension and hydrodynamic instabilities. One of the most prominent examples is viscous fingering in quasi-two-dimensional Hele-Shaw geometries, where the displacement of a more viscous fluid by a less viscous one produces highly branched, dendritic morphologies. In this work, we report the formation of nearly dendritic patterns in Langmuir Reverse Schaefer ($\mathcal{L}$ -RSh) films of the nematic liquid crystal *N*-(4-methoxybenzylidene)-4-butaniline (MBBA) upon incubation with silver nanocolloids (AgNcs) of average diameter ~ 30 nm, whereas linear stripelike structures are observed for larger nanoparticles (~ 64 nm) on the same $\mathcal{L}$ -RSh film of MBBA molecules. The origin of this size-dependent morphological transition is interpreted in terms of topological defects generated due to the incorporation of metal nanoparticles within the continuum $\mathcal{L}$ -RSh liquid crystal matrix. A comprehensive statistical characterization of the patterned substrate has been carried out through parameters such as lateral correlation length (ξ), fractal dimension (D), Hurst exponent (β), and interface width (ω). Further insights into the complexity of the patterns reveal their multifractal nature, evaluated using two-dimensional multifractal detrended fluctuation analysis. To the best of our knowledge, this study appears to be the first report where thorough investigations on the surface properties of the as-prepared patterned substrate have been discussed in detail. Finally, the patterned substrate demonstrates the good performance as surface-enhanced Raman scattering platforms, enabling detection of rhodamine 6G and 1,10-phenanthroline molecules at extremely low concentrations.

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I. INTRODUCTION

Spontaneous formation of patterns at fluid-fluid interfaces is a ubiquitous phenomenon in nonequilibrium systems [1–3]. One of the most well-known examples is the viscous fingering instability, where a less viscous fluid displaces a more viscous one in quasi-two-dimensional Hele-Shaw cells [4–6]. The resulting tip splitting, fingerlike or highly branched structures are known to emerge from the interplay between interfacial tension and the hydrodynamic instability emerging out due to the viscosity contrast between the two fluids [7–9]. Similar natures of fascinating pattern

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formations are also seen in liquid crystals (LCs) and nanoparticle composites that have few reports and largely remain unexplored [10–13].

LCs and metal nanoparticles are two fascinating classes of materials that have generated significant interest in recent times both in fundamental research and in applied research nowadays [14–17]. LC molecules have long-range orientational ordering with compromised positional ordering characterized by their order parameters. LCs are extensively utilized in technologies such as liquid crystal displays, light shutters, sensors, and modulators [18–21]. In contrast, distinctive surface plasmon resonance characteristics of metal nanoparticles mostly in the visible region of the electromagnetic spectrum have enabled their broad applications in plasmonics, metal-enhanced fluorescence, and also in surface-enhanced Raman scattering (SERS) studies [22–24]. Considering these things in mind, here we report formation of surprising dendritic patterns in Langmuir Reverse Schaefer ($\mathcal{L}$ -RSh) films of *N*-(4-methoxybenzylidene)-4-butaniline (MBBA) upon incubation in silver nanocolloid (AgNc) solution of average diameter ~ 30 nm, although the same $\mathcal{L}$ -RSh film shows linear stripes for AgNcs with larger average diameter (~ 64 nm). The reason behind such pattern formation is not the same as observed in Hele-shaw cells; instead, it has been discussed in terms of topological defects that arise due to inclusion of AgNcs in the continuum $\mathcal{L}$ -RSh film of LCs. To have a complete panorama about the microscopic features of the patterns, the surface properties of LC-nanoparticle composite on the $\mathcal{L}$ -RSh film matrix have been elaborately discussed using statistical parameters such as lateral correlation length (ξ), fractal dimension (D), the Hurst exponent (β), and interface width (ω). Deeper analyses of the fractal features linked with the patterns induced on the surface reveal their multifractal nature. Thus, its multifractal nature has also been explored from two-dimensional multifractal detrended fluctuation analysis (2D-MFDFA). However, the patterned domains reported so far for similar systems [Langmuir-Blodgett (LB)/ $\mathcal{L}$ -RSh films, LC-nanoparticle composite] prior to this are distinctively dissimilar from the observed stripe and dendritic features as reported here [11,25–27]. Thus, to the best of our knowledge, the specific linear stripe- and dendriticlike features as observed in the case of the present substrates upon inclusion of silver nanoparticles (AgNps) of varied sizes (ca. 64 and 30 nm) in $\mathcal{L}$ -RSh film of MBBA are observed for the first time. Furthermore, to the best of our knowledge, the multifractal features of the substrate have also been reported for the first time in detail from 2D-MFDFA, albeit evidence of the fractal nature of the Langmuir monolayers and in LB films of stearic acid dipped in silver colloid is reported elsewhere [28,29]. Finally, the substrate has been shown to act as efficient SERS sensing platform for the first time that can detect rhodamine 6G (R6G) and 1,10-phenanthroline (Phen) molecules at ultralow concentrations ($\sim 10^{-12}$ and 10^{-10} M). A comparison table with prior LC-nanoparticle, LB/ $\mathcal{L}$ -RSh, and SERS substrates is shown in Table S1 of the Supplemental Material [30], along with the corresponding references [10,11,25–27]. We believe that such patterned substrate will enable one to spot the hot spots very easily using Raman microscope, which in turn will exhibit optimized SERS signals useful for sensing and in diagnostics.

II. EXPERIMENTAL PROCEDURE

A. Materials and methods

Spectroscopic-grade MBBA, LC, R6G, and Phen were procured from Sigma-Aldrich and used without further purification. Silver nitrate (AgNO_3), trisodium citrate (TSC), sodium hydroxide, sodium borohydride (NaBH_4), hydrogen peroxide, sulfuric acid, ethanol, spectroscopic-grade chloroform, and acetone were obtained from E. Merck (Germany). All glass and quartz slides used in the synthesis were thoroughly cleaned with freshly prepared aqua regia and piranha solution. The silicon wafers used in the experiments were cleaned with chloroform and acetone. Subsequently, the cleaned slides and wafers were rinsed several times with triple-distilled deionized water (Milli-Q Plus system, Millipore Corporation, USA) having an electrical resistivity of 18.2 M Ω cm and a pH of approximately 6.8. The same deionized water was used throughout for the preparation of all required solutions.

The silver nanocolloids of mean particle size ~ 64 and 30 nm (AgNc1 and AgNc2) were synthesized following the modified Lee-Meisel method, as reported earlier [31]. Briefly, AgNc1 and AgNc2 were synthesized by reducing aqueous solution of AgNO_3 using NaBH_4 and TSC as reducing agents. Fresh aqueous solutions of NaBH_4 and TSC were mixed and heated at 60°C for 30 min under vigorous stirring in the dark. Thereafter, the required volume of AgNO_3 solution was incorporated dropwise to the separate sets of NaBH_4 and TSC mixtures, and the temperature was increased to 90°C . The pH of the mixtures was adjusted to 10.5 using 0.1 M NaOH . The reaction was continued until the initially colorless solutions gradually changed to pale and bright yellow-green colors indicating the formation of AgNc1 and AgNc2. The as-prepared colloids were cooled to room temperature and centrifuged for 15 min to remove unreacted species. The final AgNc1 and AgNc2 colloids were stored at 4°C for subsequent use. The average particle size of the AgNps can be modified by simply varying the respective concentrations of AgNO_3 , NaBH_4 , and TSC. The UV-Vis spectra of the as-prepared AgNc1 and AgNc2 colloidal solutions show characteristic plasmon resonance peaks centered at ~ 430 and 410 nm, respectively. The zeta (ζ) potentials of AgNc1 and AgNc2 are estimated to be ~ -33.5 and -38.4 mV, respectively. These results indicate that the as-prepared colloids (AgNc1 and AgNc2) are highly stable. The concentration of Ag for AgNc1 and AgNc2 has been estimated to be $\sim 7.25 \times 10^{11}$ and 1.71×10^{13} per ml. The colloidal stabilities have been checked from ζ potential measurements after 2 months of their synthesis. The ζ values are estimated to be ~ -30.9 and -31.7 mV for AgNc1 and AgNc2, respectively, which indicate that the as-prepared colloids are highly stable even after 2 months. Furthermore, ζ potential measurements of AgNc1 and AgNc2 exhibit single peak in the particle size distribution plot [Figs. S2(c) and S2(d) of the Supplemental Material [30]]. The polydispersity index for AgNc1 and AgNc2 has been estimated to be ~ 0.189 and ~ 0.188 , indicating that both the colloids are nearly monodisperse [32,33]. These results are further substantiated by the TEM histogram [Figs. S2(a) and S2(b) of the Supplemental Material [30]] plots whose distributions show nearly monodisperse nature of both the silver nanocolloids with mean sizes ~ 64 and 30 nm and corresponding standard deviations (σ) ~ 5.9 and 3.5 , respectively [34].

The room-temperature surface pressure–area (π – A) isotherm of MBBA was recorded using a computer controlled dual LB trough (Model D2007, Apex Instruments). The LB trough subphase consisted of triple-distilled deionized water maintained at near-neutral pH (~ 6.8). Surface pressure measurements were performed by applying filter paper as the Wilhelmy plate. Approximately $200\ \mu\text{l}$ of a 10^{-4} M MBBA solution in chloroform was spread dropwise onto the water surface using a microsyringe. After allowing the solvent to evaporate for 20 min, the Teflon barriers were compressed at a constant rate of $2\ \text{mm/min}$. The hysteresis behavior of the molecules during successive compression-expansion cycles of the π – A isotherms has been investigated and shown in Fig. S4(a) of the Supplemental Material [30]. The successive compression and expansion cycles closely retrace the same path in the π – A isotherm, indicating negligible hysteresis loss for MBBA molecules at the air-water interface. These observations suggest that the molecular conformation remains stable during repeated cycling and demonstrates the excellent reversibility and reusability of the film.

The $\angle$ -RSh films of MBBA molecules were transferred onto solid substrate at a surface pressure of $20\ \text{mN/m}$, with the vertical lifting speed fixed at $2\ \text{mm/min}$ at room temperature. The $\angle$ -RSh technique for lifting LB films was initially proposed by our research group whose details are available in our earlier publications [10,35]. The transfer ratio and the measured $\angle$ -RSh film thickness from atomic force microscope (AFM) [Figs. S4(b) and S4(c) of the Supplemental Material [30]] are estimated to be ~ 0.9 and $1.5\ \text{nm}$ for a single deposited layer.

The uplifted $\angle$ -RSh films of MBBA were then dipped in AgNc1 and AgNc2 colloidal solutions for $48\ \text{h}$. The films were then withdrawn slowly, dried in a hot-air oven, and stored for further characterization. The $\angle$ -RSh films of MBBA treated with AgNc1 and AgNc2 are hereafter referred to as the substrate A (S_A) and substrate B (S_B). The elemental composition of the as-prepared substrates has been confirmed by energy-dispersive x-ray spectrometry (EDS) mapping, elemental line scans, and x-ray

photoelectron spectroscopy (XPS), as shown in Fig. S5 of the Supplemental Material [30]. The corresponding analyses reveal the presence of silver, oxygen, nitrogen, and carbon on the as-prepared substrates.

B. Instrumentations

The absorption spectra were obtained using a Jasco UV-Vis absorption spectrometer (model: V-630). The SERS spectra were measured with a NEW XploRA Plus V1.2 Å MULTILINE Confocal Raman microscope (model: XPLOA+/ML), equipped with a 1800 groove/mm holographic grating and a TE air-cooled HORIBA Scientific charged-coupled device detector. The samples were excited with a 532 nm green solid-state laser, with a spot size of 0.7 μm in diameter and a laser power of 0.4 mW. Each spectrum was recorded with a data acquisition time of 10 s. The scattered signals were collected in a backscattering geometry from the excitation using an Olympus open-stage microscope with a 100 $\times$ objective. The detector and associated equipment were controlled via LabSpec 6 software provided by Horiba. Polarized optical microscopic (POM) images of the as-prepared substrates were taken from Leica DM 2700P polarizing microscope. The surface morphology of the substrates was examined with a field emission scanning electron microscope (FESEM) (Inspect FEI F50). The EDS analyses of the substrates were performed using a Quanta FEG 250. The surface topology of the as-prepared substrates was characterized using AFM (Asylum Research, model No. MFP-3D) operated in tapping mode. XPS measurements were carried out using Omicron XPS fitted with a monochromated Al K α x-ray source. The ζ potentials and hydrodynamic diameters of the as-synthesized AgNcs were determined by dynamic light scattering (DLS) measurements using a NanoZS (Malvern) instrument.

C. Theoretical details

1. Estimation of statistical parameters linked with the topography of the substrate

The statistical parameters associated with the topographical characteristics of the as-prepared substrate have been calculated from the corresponding AFM image. The lateral correlation lengths (ξ), which describe the correlation of the surface heights at any two points on the substrate, have been estimated from the autocorrelation function $R(r_1, r_2)$ [36,37]. Accordingly,

$$R(r_1, r_2) = \frac{G(r_1, r_2)}{C^2}, \quad (1)$$

where $G(r_1, r_2)$ is represented as autocovariance function expressed as [36,37]

$$G(r_1, r_2) = E\langle h(r_1)h(r_2) \rangle = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} h_1 h_2 P_j(h_1, h_2; r_1, r_2) dh_1 dh_2, \quad (2)$$

where h_1, h_2 represent the heights at two different positions on the surfaces r_1, r_2 , respectively, $P_j(h_1, h_2; r_1, r_2)$ corresponds to joint distribution function, and C^2 indicates the autocovariance function when r_1 and r_2 are merged, i.e., $|r_1 - r_2| = 0$. For rough surfaces, both the autocovariance function $G(r_1, r_2)$ and the autocorrelation function $R(r_1, r_2)$ depend only on the separation distance r between the two positions on the substrate surface. Therefore, these functions can be written in simplified form as

$$G(r_1, r_2) = G(|r_1 - r_2|) = G(r), \quad R(r_1, r_2) = R(r). \quad (3)$$

The topographical characteristics of the substrate obtained from the AFM image have been further estimated from height-height correlation function (HHCF). The HHCF is represented as [36,37]

$$H(r) = 2\omega^2 [1 - e^{-(\frac{r}{\xi})^{2\beta}}] = 2\omega^2 f\left(\frac{r}{\xi}\right), \quad (4)$$

where r denotes the displacement vector, ω^2 represents the mean-square surface roughness, β is the Hurst exponent (or roughness exponent), and ξ corresponds to the lateral correlation length. The function $f(x)$ is the scaling function of $f(\frac{r}{\xi})$, which exhibits the following relations:

$$f\left(\frac{r}{\xi}\right) = f(x), \quad f(x) = x^{2\beta}, \quad \text{for } x \ll 1, \quad f(x) = 1, \quad \text{for } x \gg 1. \quad (5)$$

The Hurst exponent (β) characterizes the short-range roughness of the surface in the regime $r \ll \xi$ and ω denotes the root-mean-square roughness or interface width for $r \gg \xi$. Thus, the interface width represents its long-range roughness characteristics of the surface.

The geometric complexity of the patterned substrate has also been estimated from the contour fractal dimension using the box-counting method following the relation [38–40]

$$N(s) \propto s^{-D_c}, \quad (6)$$

where D_c is the contour fractal dimension, s represents the box size, and $N(s)$ denotes the number of boxes. Image processing workflow used for fractal analysis is shown in Fig. S9 of the Supplemental Material [30].

More general multifractal nature of the substrate as evaluated from fractal dimension calculation has been explored from 2D-MFDFA. A fractal is simply a geometric pattern that repeats itself in nearly the same way across all scales. It can be divided into two groups, such as monofractals and multifractals. The same scaling characteristics of the recurring patterns at various system regions define a monofractal system. On the other hand, a multifractal system requires a large number of scaling exponents for accurate characterization, or it is made up of fractals with varying weights. MFDFA is the source of the MF approach. Kantelhardt *et al.* [41] first developed the MFDFA method as a generalization of detrended fluctuation analysis [42]. The beauty of multifractal analysis is that different scaling exponents are used for fractal subsets, which are differently interlaced. Simple monofractal scaling, which can be characterized by a single exponent, is not evident in many systems. In certain instances, crossover scales are present, but in other cases, scaling behavior was discovered to be much more complex, requiring distinct exponents for each series segment. Thus, the introduction of a multifractal formalism seemed appropriate. In time series, multifractality generally comes in two varieties. These are the multifractality resulting from a wide probability density function in relation to the time series values and the other caused by different long-range correlations between small and big fluctuations. Due to its superior accuracy and reduced computational expense, the MFDFA method has demonstrated its efficacy across numerous fields, including human gait, economic time series, earthquake signals, and heart rate dynamics. Even though the technique was created to investigate time series fluctuation, it has also been successfully used to examine the multifractal behavior in spatial pattern [43]. Moreover, MFDFA has been extensively used to analyze SEM/AFM pictures and has produced useful results for studying the surface morphology of various kinds of substrates [44]. Here, the multifractal behavior of the AgNps impregnated in the $\mathcal{L}$ -RSh film of MBBA molecule has been analyzed from their respective FESEM images using the 2D-MFDFA method. Four independently prepared FESEM images having size of 1024×943 pixels have been acquired. Representative regions of interest (ROIs) have been selected from each FESEM image. A total of 47 nonoverlapping image patches, each measuring 120×120 pixels, have been extracted from these ROIs for multifractal analysis. The minimum patch size required for the application of 2D-MFDFA is 100×100 pixels [45,46]. Prior to analysis, all image patches have been converted to grayscale to preserve intensity variations and subsequently analyzed using the 2D-MFDFA method. The steps of 2D-MFDFA are listed below [45,46]:

Step 1: Consider an image represented by a two-dimensional array $I(i, j)$, where $i = 1, 2, \dots, M_1$ and $j = 1, 2, \dots, M_2$. The array is partitioned into $M_{1s} \times M_{2s}$ nonoverlapping square segments of equal size $s \times s$, where $M_{1s} = \lfloor M_1/s \rfloor$ and $M_{2s} = \lfloor M_2/s \rfloor$. Each segment, denoted by $I_{(c,d)}$, is defined as $I_{(c,d)}(i, j) = I(p_1 + i, p_2 + j)$ for $1 \leq i, j \leq s$, where $p_1 = (c - 1) \times s$ for $c = 1, 2, \dots, M_{1s}$ and $p_2 = (d - 1) \times s$ for $d = 1, 2, \dots, M_{2s}$.

In the present work, $I(i, j)$ denotes the analyzed FESEM image patches.

Step 2: The cumulative sum $T_{c,d}(i, j)$ for each segment $I(i, j)$ is computed as follows:

$$T_{c,d}(i, j) = \sum_{u=1}^i \sum_{v=1}^j I_{c,d}(u, v), \quad (7)$$

where $1 \leq i, j \leq s$.

Step 3: The trend of the generated $T_{c,d}$ can be characterized by fitting it with different bivariate polynomial functions [47]. The simplest planar function has been employed in the present study [48,49]:

$$\widetilde{T}_{c,d}(i, j) = ai + bj + k, \quad (8)$$

where $1 \leq i, j \leq s$ and a, b , and k denote the free parameters obtained by applying the least-squares method via matrix operations. The residual matrix can be written as

$$\varepsilon_{c,d}(i, j) = T_{c,d}(i, j) - \widetilde{T}_{c,d}(i, j). \quad (9)$$

The detrended fluctuation function $F_{c,d}(s)$ of the segment $I_{c,d}$ is calculated from the variance of the residual matrix $\varepsilon_{c,d}(i, j)$ and is expressed as follows:

$$F_{c,d}(s) = \sqrt{\frac{1}{s^2} \sum_{i=1}^s \sum_{j=1}^s (\varepsilon_{c,d}(i, j))^2}. \quad (10)$$

Step 4: Subsequently, the overall detrended fluctuation function is obtained by averaging the detrended fluctuation functions over all the segments, as expressed below:

$$F_q(s) = \left\{ \frac{1}{M_{1s} \times M_{2s}} \sum_{c=1}^{M_{1s}} \sum_{d=1}^{M_{2s}} F_{c,d}^q(s) \right\}^{\frac{1}{q}}, \quad (11)$$

where q may assume any real value except for $q = 0$.

When $q = 0$, we have

$$F_o(s) = \exp \left\{ \frac{1}{M_{1s} \times M_{2s}} \sum_{c=1}^{M_{1s}} \sum_{d=1}^{M_{2s}} \ln[F_{c,d}(s)] \right\}. \quad (12)$$

Step 5: By varying the value of s in the range from $s_{\min} \approx 6$ to $s_{\max} \approx \min[M_1, M_2]/4$, the scaling relationship between the detrended fluctuation function $F_q(s)$ and s can be determined using the following relations:

$$F_q(s) \propto s^{h(q)}, \quad (13)$$

where $h(q)$ denotes the generalized Hurst exponent. For each value of q , the multifractal scaling exponent is obtained as follows:

$$\tau(q) = qh(q) - 2. \quad (14)$$

The singularity exponent $\alpha(q)$ and the singularity spectrum $f(\alpha)$ are estimated as

$$\alpha = \frac{d\tau(q)}{dq}, \quad (15)$$

$$f(\alpha) = q[\alpha - h(q)] + 2. \quad (16)$$

The plot of $f(\alpha)$ versus α is referred to as the multifractal spectrum.

Spectrum width $\Delta\alpha$ is the difference between maximum ($\alpha_{\max}$) and minimum ($\alpha_{\min}$) values of singularity exponent α . Higher $\Delta\alpha$ indicates stronger multifractality of the signal. For a monofractal series, the spectrum collapses to a single point and the width of the spectrum will be zero. Third

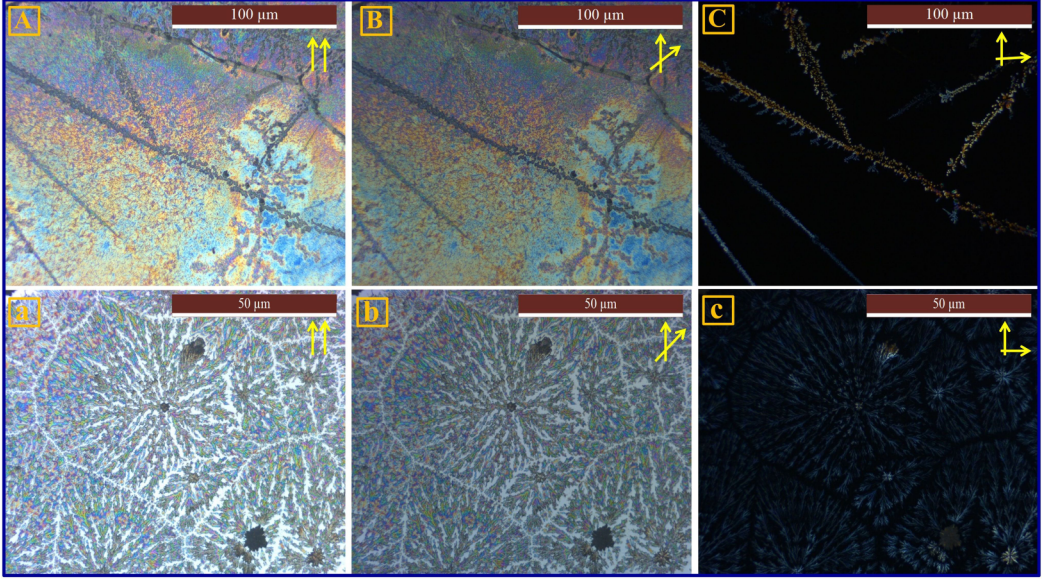


FIG. 1. POM images of S_A [panels (A)–(C)] and S_B [panels (a)–(c)]. (Left, middle, and right columns denote that the images are taken under parallel, slightly crossed, and crossed conditions.)

feature α_0 is equal to the value of α at $q = 0$. Maximum value of $f(\alpha)$ is 2 for 2D signal and is found when $q = 0$. So, $f(\alpha)$ is maximum at α_0 . Higher value of α_0 signifies higher correlation and regularity of the analyzed signal. Shape of multifractal spectrum can be described by asymmetry parameter A_α [46]:

$$A_\alpha = \frac{\Delta\alpha_L - \Delta\alpha_R}{\Delta\alpha_L + \Delta\alpha_R}, \quad (17)$$

where $\Delta\alpha_L = \alpha_0 - \alpha_{\min}$ and $\Delta\alpha_R = \alpha_{\max} - \alpha_0$. For a symmetric multifractal spectrum, A_α becomes zero. A_α is positive, signifying a right-skewed spectrum, whereas $A_\alpha < 0$ is associated with a left-skewed spectrum.

The spatial distributions of electric fields around the plasmonic nanostructures have been investigated using three-dimensional finite difference time domain (3D-FDTD) simulations. The optical properties of silver have been described using the experimentally measured Johnson-Christy dielectric data, while water with a refractive index of 1.33 has been considered as the surrounding medium. The nanostructure models have been illuminated with a linearly polarized plane wave (PW) having an electric-field amplitude of 1.0 V/m at an incident wavelength of 532 nm. The simulation duration has been fixed at 500 fs to ensure stable and converged electric field distributions.

III. RESULTS AND DISCUSSIONS

A. POM, FESEM, and AFM images of S_A and S_B

POM images of pristine LCs and the nanoparticle composites provide important information about intrinsic and induced topological defects [50–52] in the system. These defects arise from broken continuous symmetry and strongly influence the spatial orientation of the LC director field on the film surface. Such defect sites can also act as trapping centers for nanoparticles, promoting their unusual self-assembly [53,54]. The POM images of substrates S_A and S_B are shown in Fig. 1.

Under parallel polarization, S_A shows dark linear stripelike features within birefringent domains along with fractal-like microaggregates near the edges. Slightly crossed polarization does not

produce major changes [Fig. 1(B)]. However, under fully crossed polarizers [Fig. 1(C)], the birefringent background disappears, resulting in a dark field with bright linear stripes. The dark background primarily indicates homeotropic alignment of MBBA molecules, while the bright linear defects suggest interactions between the LC matrix and AgNps [55,56].

In contrast, S_B exhibits distinctly different textures [Figs. 1(a)–1(c)]. Under parallel polarization, multiple radially outward defect structures emerge from several source points and extend over micrometer length scales. Under crossed polarization, these defects appear as bright radial traces within a dark LC background [Fig. 3(c)]. Detailed observation reveals slight distortions along the radial direction along with branching, dendriticlike features. Such morphologies likely arise from interfacial instabilities caused by increased anisotropy in the system, possibly influenced by constraints from the LB trough barrier and/or the presence of AgNps [9,57–59].

Incorporation of AgNps into the $\angle$ -RSh matrix of MBBA liquid crystals disturbs the orientational ordering of LC molecules, which may induce elastic distortions as guided by the Frank free energy. These distortions in turn may be counterbalanced by the surface anchoring energy, which tends to realign the LC director along preferred easy axes. Spatial distribution of the Frank director on the substrate is perhaps decided by the mutual trade-off between elastic ($F_E \sim Ka$, where K is elastic constant and a is the particle size) [60] and surface anchoring energies ($F_S \sim Wa^2$, where W is surface energy term and a is the particle size) [60], which in turn depend on the size (a) and square of the size (a^2) of the nanoparticles, respectively.

For larger aggregated AgNps (average diameter ~ 64 nm) on the S_A substrate, surface anchoring energy (F_S) dominates over F_E , which promotes homeotropic alignment of MBBA molecules around the nanoparticles. This may lead to the formation of radial hedgehogs accompanied by their corresponding hyperbolic hedgehog counterparts (to satisfy topological charge conservation criterion for infinite boundary condition), forming dipolar defect structures that appear as linear stripes in POM images [11,61].

For smaller aggregated AgNps (average diameter ~ 30 nm) on the S_B substrate, F_E seems to be dominant over F_S , and may favor the formation of surface ring defects [62,63]. However, both S_A and S_B substrates exhibit energetically unfavorable defect cores with reduced orientational order. To minimize the free energy associated with these topological defects, AgNps (which themselves are responsible for creating defects) move toward the defect zones as guided by the strongest gradient of the Frank directors.

Alternatively, surface patterns as seen on the substrates may also originate from drying artifacts, coffee-ring effects, dewetting, capillary flow, or substrate-induced aggregation. To nullify their contributions, a series of control experiments have been performed. The control experiments are accomplished from pristine MBBA $\angle$ -RSh films, MBBA films immersed in water without AgNcs, AgNcs deposited on bare silicon substrates, dried AgNcs without MBBA, and films incubated for 24, 48, and 72 h. The corresponding optical microscopic images are shown in Fig. S3 of the Supplemental Material [30]. For MBBA $\angle$ -RSh-AgNc1 system, while discernable linear stripes are clearly seen for the substrate dipped in AgNc1 for 48 h [Fig. S3(h) of the Supplemental Material [30]], prominent complex networklike features are recorded for the same dipped in AgNc1 for 72 h [Fig. S3(i) of the Supplemental Material [30]]. However, for MBBA $\angle$ -RSh-AgNc2 system, clear and discernible dendritic domains are only seen for the substrate dipped in AgNc for 48 h [Fig. S3(k) of the Supplemental Material [30]]. The experimental results collectively demonstrate that the patterns on the substrates are primarily driven by the interplay between elastic and surface anchoring energies as discussed earlier.

To check the morphological patterns (as being envisaged from POMs), the FESEM images of both the substrates have been recorded. The corresponding FESEM images of S_A and S_B under different scales of magnification are shown in Fig. 2.

Both the images replicate the respective POM images as shown in Fig. 1 and clearly indicate impregnations of AgNps within the defect domains. While S_A substrate exhibits elongated, well-defined stripelike structures ($\sim 200 \mu\text{m}$ in length and $\sim 4 \mu\text{m}$ in width) emerging from the two-dimensional surface, S_B , on the other hand, shows radially expanding particle-rich domains

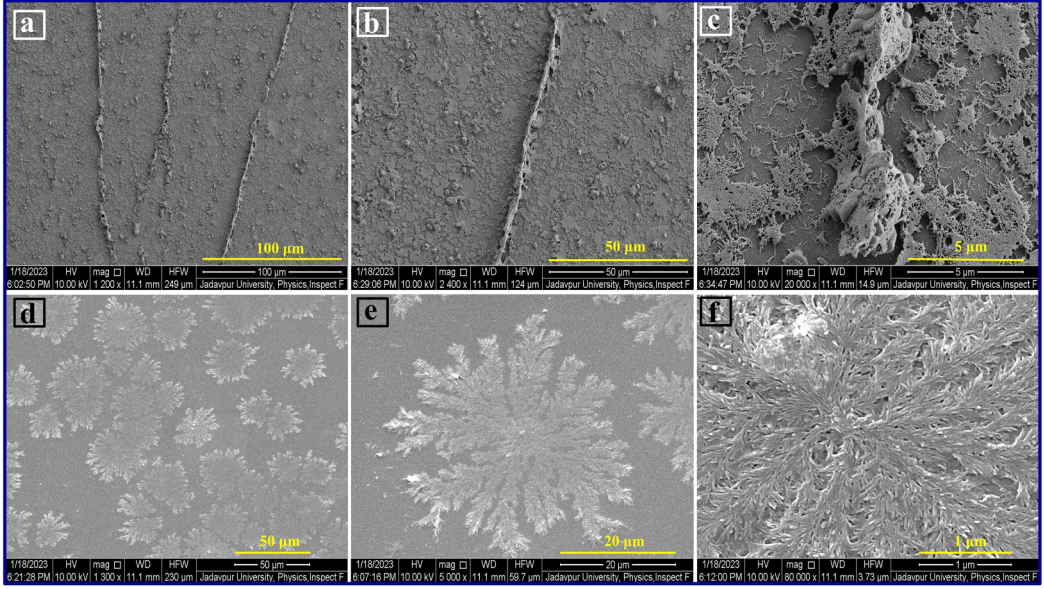


FIG. 2. FESEM images of S_A [panels (a)–(c)] and S_B [panels (d)–(f)] under different scales of magnifications.

($\sim 20 \mu\text{m}$ radius) originating from multiple origins. To further examine the morphological patterns observed in the POM and FESEM images, AFM images of both substrates have been recorded. The corresponding 2D and 3D AFM images of the S_A and S_B substrates are shown in Figs. S4(d)–S4(g) of the Supplemental Material [30]. The observed surface morphologies are in good agreement with the features revealed by the POM and FESEM images, thereby confirming the consistency of the structural characteristics across the different characterization techniques.

For direct evidence of AgNps within defect domains, the EDS mapping, elemental line scans of the S_A and S_B substrates have been recorded. The corresponding results are shown in Fig. S5 of the Supplemental Material [30]. The elemental maps of S_A and S_B confirm the presence of aggregated AgNps within defect domains, consistent with the observations from the POM images. Such clustered AgNps assemblies are highly favorable for the generation of plasmonic hot spots. To demonstrate the reproducibility of the observed surface morphologies, additional FESEM images of the S_A and S_B substrates are presented in Figs S2(e) and S2(f) of the Supplemental Material [30].

The overall images of the substrates indicate that in comparison to S_A , S_B apparently shows fractal or probably multifractal-like patterns [Figs. 1(a)–1(c) and 2(d)–2(f)], which demand its detail analyses of topographical features as discussed in the next section.

B. Topographical features of the S_B substrate

The statistical parameters associated with the topographical features of the S_B have been calculated from the corresponding AFM image using autocorrelation function $[R(r)]$ and the HHCF.

The value of ξ , which reflects the degree of correlation of surface height between two points on the substrate, has been determined from the autocorrelation function $R(r)$. The variation of the autocorrelation function $R(r)$ with the displacement vector $\vec{r}$ for S_B is presented in Fig. 3(a). The value of ξ is obtained from the position where $R(r)$ decreases to $1/e$ of its maximum value at zero lag (i.e., at $r = |r_1 - r_2| = 0$). For S_B , the ξ value is estimated to be approximately 176 nm. This ξ value indicates that the surface features remain spatially correlated over a nanoscale

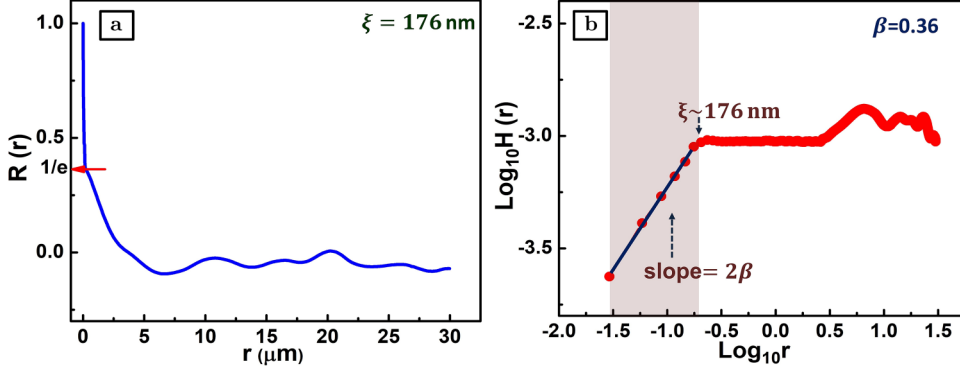


FIG. 3. (a) The autocorrelation function $R(r)$ as a function of displacement vector $\vec{r}$. (b) Variation of $\log H(r)$ as a function of $\log r$ for the S_B (here, $\log = \log_{10}$).

distance of approximately 176 nm and also corresponding to the surface roughness observed in the AFM image over which the surface-height fluctuations remain spatially correlated. Our previous reports suggest that these types of ξ values are responsible for SERS enhancement of the substrate [36,37].

Figure 3(b) presents the HHCF plot illustrating the variation of $\log_{10} H(r)$ as a function of $\log_{10} r$ plot for the substrate. The HHCF data were obtained from the AFM image of the respective substrate. Interestingly, the log-log plot exhibits a linear dependence at shorter length scales ($r \ll \xi$), indicating a power-law behavior. However, at larger length scales ($r \gg \xi$) the HHCF curves approach a saturation limit reflecting their asymptotic behavior.

The ξ value of the substrate determined from the corresponding HHCF plot is estimated to be $\sim 176 \text{ nm}$. This ξ value also suggests that the surface features remain spatially correlated over a nanoscale distance of approximately 176 nm. This value is in agreement with that obtained from the autocorrelation function. The values of β and ω from the HHCF analysis are estimated to be ~ 0.36 and 21.6 nm for the substrate. These β and ω values indicate the jagged nature and long-range roughness characteristics of the surface.

The value of β obtained from the HHCF analysis is further employed to calculate the surface fractal dimensions of the LC-nanoparticle composite on the $\mathcal{L}$ -RSh film matrix that are allocated on S_B . Surface fractal dimension (D_S) has been calculated using the following relation [37]:

$$D_S = d + 1 - \beta, \quad (18)$$

where $d + 1$ is referred to the dimension of the embedded space and β is the Hurst exponent. Considering dimension of the embedded space to be 3, the fractal dimension (D_S) of S_B has been estimated to be ~ 2.64 .

So, primarily the substrate exhibits fractal nature of the LC-nanoparticle composite on the $\mathcal{L}$ -RSh film matrix of MBBA molecules.

C. Fractal dimension calculation of S_B and different regions of S_B

Overall, the micrographs clearly demonstrate the formation of fractal-like structures across the surface of S_B . The contour fractal dimension of S_B was determined from the slope of the $\ln N(s)$ versus $\ln(1/s)$ plot. The result, shown in Fig. 4(A), indicates that the contour fractal dimension of the substrate is 1.55 ± 0.03 . This value agrees well with the morphological features observed in the AFM micrograph and confirms the geometric complexity of the S_B surface.

The contour fractal dimensions are also calculated from different regions of the substrate. The results are shown in Figs. 4(B)–4(F). The fractal dimensions so attained from different regions of

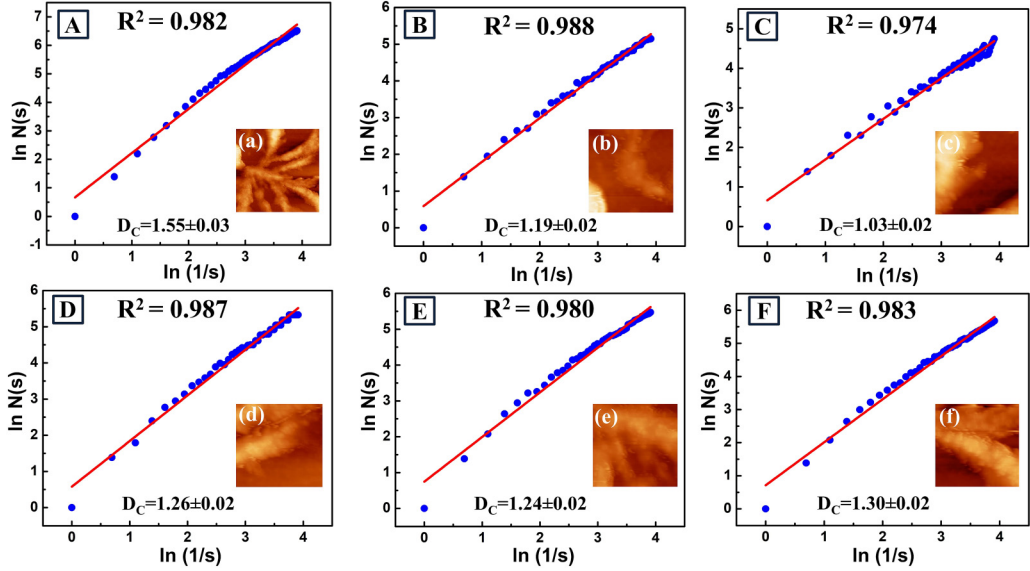


FIG. 4. $\ln N(s)$ vs $\ln(1/s)$ plots for (A) S_B and (B)–(F) for different regions of S_B . Insets: Representations of the AFM images of (a) S_B and (b)–(f) different regions of S_B .

the substrate are estimated to be in the range ~ 1.03 – 1.30 . These results suggest the substrate S_B is probably multifractal in nature. To explore the multifractal nature of the substrate, in-depth analysis has been performed.

D. Examining the topographical characteristics of S_B using 2D-MFDFA

The 2D-MFDFA have been employed to extract concealed structural information from S_B . The fluctuation functions $F_q(s)$ have been calculated for a range of “ s ,” varying from 6 to $\min[M_1, M_2]/4$ (where M_1 and M_2 denote the dimensions of the 2D surface) using Eqs. (11) and (12). These limits are commonly adopted in 2D-MFDFA studies [47,49]. The variation of $\ln F_q$ with $\ln s$ for S_B is presented in Fig. 5(a). It is observed that $\ln F_q$ versus $\ln s$ plots exhibit nearly linear behavior within the selected scale range for detrending values “ q ” varying from -10 to $+10$ in steps of 0.1 . These negative q values emphasize relatively small fluctuations, whereas positive q values emphasize larger fluctuations. This linear scaling behavior indicates the presence of long-range power-law correlations in the surface morphology. The slopes of these linear fits are used to determine the generalized Hurst exponent $h(q)$. The variation of $h(q)$ versus q is shown in Fig. 5(b) in which the value of $h(q)$ decreases with increasing q . The dependence of $h(q)$ on q of the $h(q)$ versus q plot primarily specifies the existence of multifractal characteristic of the S_B surface. Furthermore, Fig. 5(c) illustrates the nonlinear variation of the multifractal scaling exponent $\tau(q)$ with respect to q , which provides additional confirmation of the multifractal nature of the substrate. To the best of our knowledge, the results presented in Figs. 5(a)–5(c) constitute the first convincing evidence demonstrating the multifractal behavior of the S_B surface. The singularity spectrum, representing the variation of $f(\alpha)$ with the singularity exponent (α), is shown in Fig. 5(d). The spectrum shows characteristic bell-shaped curves having finite spectral widths (W). The existence of a finite W further confirms the multifractal nature of S_B , since in a purely monofractal system the singularity spectrum falls into a single point with zero width.

To evaluate the uncertainty bands of the multifractal parameters, box plots of the extracted 2D-MFDFA features, including the generalized Hurst exponent [$h(q = 2)$], multifractal scaling exponent $\tau(q = 2)$, singularity spectrum $f(\alpha(q = 2))$, spectral width (W), and asymmetry parameter

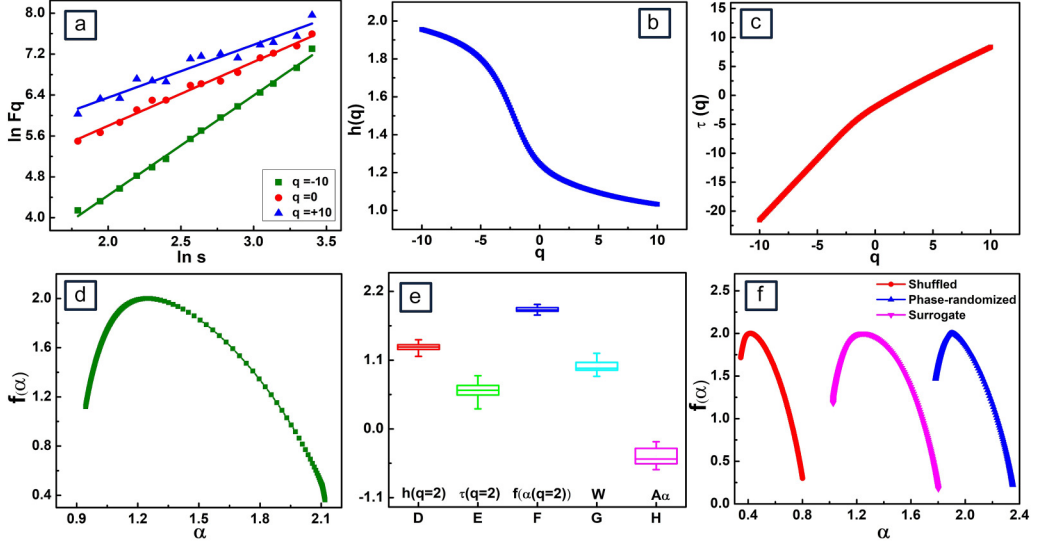


FIG. 5. Plots of (a) $\ln F_q$ vs $\ln s$ for at $q = -10, 0, +10$, (b) $h(q)$ vs q , (c) another variable $\tau(q)$ vs q , and (d) singularity spectrum $f(\alpha)$ as a function of α derived from the FESEM image of S_B . (e) Box plots depicting variation of extracted 2D-MFDFA features like $h(q = 2)$, $\tau(q = 2)$, $f(\alpha(q = 2))$, W , and asymmetry parameter A_α . (f) Singularity spectra $f(\alpha)$ as a function of α derived from shuffled, phase-randomized and surrogate images for 2D-MFDFA.

(A_α) are presented in Fig. 5(e). These distributions demonstrate the consistency and reproducibility of the multifractal characteristics across different regions of the S_B substrate. Table I presents the p values obtained from Student's t -test to evaluate whether the extracted features exhibit significant differences between the original and shuffled images. p value less than 0.05 ($p < 0.05$) indicates a statistically significant difference between the two classes. The results indicate that several extracted features differ significantly between the original and shuffled images.

To identify the possible origins of the observed multifractality, shuffled, phase-randomized and surrogate datasets have been subjected to the same 2D-MFDFA procedure. The corresponding multifractal spectrum is shown in Fig. 5(f). Random shuffling substantially reduces the spectrum width compared with the original image, indicating that the long-range correlation plays a dominant role in the observed multifractal behavior. The phase-randomized and surrogate dataset also exhibits a reduction of multifractal spectrum width than the original image. This modification demonstrates the important role of the nonlinear correlations. Overall, these analyses demonstrate that the multifractal behavior of S_B predominantly originates from long-range and nonlinear correlations.

TABLE I. p values of 2D-MFDFA features obtained from original versus shuffled image patches.

Original versus shuffled image	
$h(q = 2)$	2.748×10^{-36}
$\tau(q = 2)$	2.747×10^{-36}
W	6.290×10^{-19}

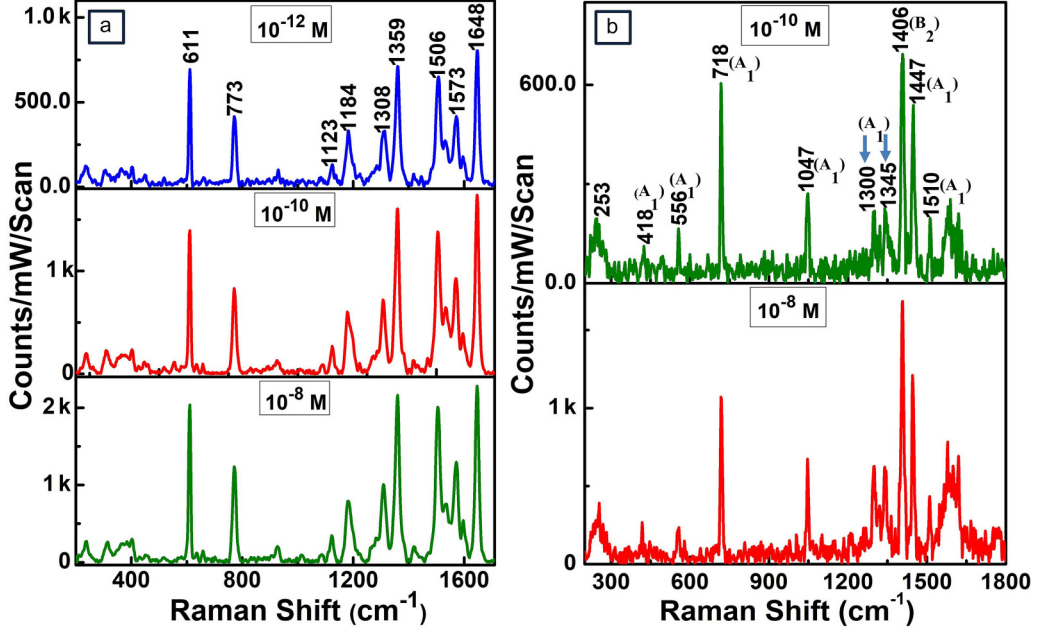


FIG. 6. SERS spectra of (a) R6G and (b) Phen molecules at different concentrations adsorbed on S_B ($\lambda_{\text{ex}} = 532$ nm).

The observed multifractal dendritic morphology, together with Ag aggregation, provides a structurally heterogeneous environment that is expected to favor the formation of electromagnetic hot spots and may be responsible to the SERS enhancement.

E. Efficacy of S_B as SERS active substrate

The SERS performance of S_B was estimated using R6G and Phen as probe molecules under on- and off-resonant conditions, respectively. Figure 6(a) presents the SERS spectra of R6G adsorbed on S_B at different concentrations ranging from 10^{-8} to 10^{-12} M. The observed results are consistent with our expectations, showing substantial enhancement of Raman signals at approximately 611, 773, 1123, 1184, 1308, 1359, 1506, 1573, and 1648 cm^{-1} [64,65]. These pronounced enhancements enable clear detection of the characteristic vibrational signatures of the probe molecules even at extremely low concentrations. On the other hand, SERS measurements of Phen were also carried out at concentrations in the range of $\sim 10^{-8}$ to 10^{-10} M. The corresponding spectra are displayed in Fig. 6(b). The SERS spectra of Phen show significantly intensified Raman bands associated with vibrational modes belonging to the A_1 and B_2 symmetry species of the molecule [66]. The enhancement factors (EFs) for Raman bands of R6G and Phen molecules have been calculated using the following relation [67]:

$$\text{EF} = \frac{I_{\text{SERS}}/N_{\text{SERS}}}{I_{\text{bulk}}/N_{\text{bulk}}}, \quad (19)$$

where I_{SERS} and I_{bulk} indicate the SERS and normal Raman (NR) intensities of the probe molecule. N_{SERS} and N_{bulk} denote the number of the probe molecule in SERS and NR measurements. N_{SERS} and N_{bulk} have been determined using the relation shown below:

$$N_{\text{bulk}} = C_{\text{bulk}} A h N_A, \quad (20)$$

$$N_{\text{SERS}} = C_{\text{SERS}} V N_A A' / A'. \quad (21)$$

Here, C_{bulk} , C_{SERS} , A , h , N_A , A' , and V denote concentration of probe at normal Raman measurement, concentration of probe at SERS measurements, laser spot area ($4 \times 10^{-13} \text{ m}^2$), confocal depth ($0.8 \text{ }\mu\text{m}$), Avogadro number, area of the probe molecule dropcasted on the slide $4 \times 10^{-5} \text{ m}^2$, and volume of the probe molecule ($25 \text{ }\mu\text{l}$). The normal Raman spectra of 0.1 M of R6G and 1 M Phen have been shown in Figs. S6(a) and S6(b) of the Supplemental Material [30]. In the concentration range 10^{-8} to 10^{-12} M , the monolayer approximation is reasonable for R6G and Phen molecules [68,69], and almost all molecules in the droplet within the laser spot area ($\sim 4 \times 10^{-13} \text{ m}^2$) contribute to SERS. The calculated EFs for the Raman bands of R6G and Phen are shown in Tables S2 and S3 of the Supplemental Material [30]. From Tables S1 and S2 of the Supplemental Material [30], it is seen that $\sim 10^4$ – 10^8 and 10^5 – 10^7 orders of enhancement have been recorded for R6G and Phen adsorbed on S_B . The analytical performance of the developed SERS substrate has been evaluated using different concentrations of the R6G molecule. The characteristic Raman band at 611 cm^{-1} has been selected as the analytical signal and its intensity has been plotted as a function of concentration to establish the calibration relationship, as shown in Fig. S7(h) of the Supplemental Material [30]. A linear calibration curve is obtained over the concentration range of 1 – 100 pM , with a correlation coefficient R^2 of 0.98 . To determine the limit of detection (LOD), measurements of the blank substrate have been performed under identical experimental conditions. The standard deviation of the blank response is estimated to be ~ 1.96 from independent blank measurements. The LOD is subsequently estimated using the conventional formula [64]:

$$\text{LOD} = \frac{3\sigma}{s}, \quad (22)$$

where σ and s represent the standard deviation of the blank samples and the slope of the calibration curve, respectively. The calculated LOD is 0.88 pM . These results demonstrate the concentration-dependent response, linear dynamic range, and detection capability of the developed SERS substrate. Therefore, the analytical performance is supported by quantitative calibration and statistical validation rather than solely by the observation of Raman signals of the analyte at trace concentrations. The reproducibility of the fabricated SERS substrate has been evaluated by recording SERS spectra of R6G from multiple randomly selected spots across the S_B substrate as shown in Fig. S7(a) of the Supplemental Material [30]. The intensities of the characteristic Raman band at 611 cm^{-1} are used to construct the corresponding intensity histogram as shown in Fig. S7(b) of the Supplemental Material [30]. The relative standard deviation (RSD) value is 7.9% , which indicates good reproducibility of the SERS response. To evaluate substrate to substrate reproducibility, SERS measurements are subsequently performed on multiple independently prepared S_B substrates under identical experimental conditions. The corresponding SERS spectra and characteristic Raman peak at 611 cm^{-1} intensities are shown in Figs. S7(c) and S7(d) of the Supplemental Material [30], with an RSD of 8.13% . Furthermore, independently prepared batches are examined under the same measurement conditions to assess batch to batch reproducibility. The respective SERS spectra and intensity histogram obtained from the different batches are shown in Figs. S7(e) and S7(f) of the Supplemental Material [30], with an RSD of 8.62% , which indicates good reproducibility of the substrate fabrication process. These results indicate that the substrate provides a consistent SERS response not only at different measurement positions but also among independently fabricated substrates and preparation batches for SERS measurements. To evaluate the stability of the S_B substrate, the fabricated substrates were stored under ambient conditions in open air for 1 , 2 , and 3 months. After each storage period, SERS spectra of R6G (10^{-12} M) were recorded under identical experimental conditions. The corresponding spectra are presented in Fig. S6(c) of the Supplemental Material [30]. A decrease in the Raman intensity at 611 cm^{-1} of approximately 4.7% , 10.5% , and 26.5% is observed after 1 , 2 , and 3 months of storage, respectively. The S_B substrate retained a substantial SERS response after 3 months, indicating good stability under ambient conditions. To assess the practical relevance of the present SERS substrate, its performance has been compared with representative Ag-based SERS substrates, ordered plasmonic arrays, nanoporous Ag films, Ag nancone arrays, and LC-assisted SERS substrates. The corresponding comparison is shown in

Table S4 of the Supplemental Material [30], along with the corresponding references [64,70–73]. Overall, these results demonstrate the high efficiency of the S_B substrate as a promising SERS-active platform, highlighting its potential for sensitive molecular detection and future sensing applications.

F. Near-field distributions of the aggregated AgNPs on the S_B substrate

Near-field distributions around the nanoaggregates on the S_B substrate are calculated using 3D-FDTD simulations. The simulation geometries are constructed based on the corresponding regions of the FESEM image and the simulated field distributions are presented in Fig. S8 of the Supplemental Material [30]. The simulations reveal intense electric-field localization within the nanogaps of the Ag nanoaggregates, corresponding to SERS active “hot spots.” Using the PW approximation, the corresponding enhancement factor is estimated to be $\sim 3.9 \times 10^5$. Although this value is lower than the experimentally estimated average EFs $\sim 10^8$, the difference may arise from the simplified FDTD geometry and PW approximation, as well as additional contributions from molecular resonance and charge-transfer effects. Nevertheless, the pronounced electric-field localization in the nanogaps supports the role of Ag aggregation in generating SERS hot spots and enhancing the observed SERS response.

IV. CONCLUSION

The present work demonstrates the spontaneous emergence of distinct morphological patterns in $\angle$ -RSh films of MBBA liquid crystals upon incorporation of AgNc. A size-dependent transition morphology has been observed. AgNc with an average diameter of ~ 30 nm generates dendritic patterns, whereas larger nanoparticles (~ 64 nm) produce predominantly linear stripelike domains on the same LC matrix. In contrast to classical viscous fingering instability in Hele-Shaw geometries, the observed pattern formation mechanism may be attributed to defect-mediated self-organization arising from the disruption of the orientational order of the liquid crystal host by embedded nanoparticles. The presence of topological defects introduces elastic distortions in the director field, which act as preferential trapping centers for nanoparticles and guide their spatial organization. Surface properties of the patterned substrate have been performed using statistical parameters such as the lateral correlation length (ξ), fractal dimension (D), Hurst exponent (β), and interface width (ω), respectively. Detailed examination of the fractal characteristics associated with the surface patterns indicates the presence of multifractal behavior. Furthermore, 2D-MFDFA reveals the multifractal nature of the generated surface morphologies. Such multifractal characteristics provide deeper insight into the pattern nature of structure formation in LC-nanoparticle composite films prepared via the $\angle$ -RSh technique. Finally, the patterned substrate exhibits the SERS performance, enabling detection of R6G and Phen molecules at trace concentrations up to $\sim 10^{-12}$ and $\sim 10^{-10}$ M, respectively. It may be presumed that such patterned substrate will facilitate simple identification of plasmonic hot spots using a Raman microscope, thereby enabling SERS responses for sensing and diagnostic applications.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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