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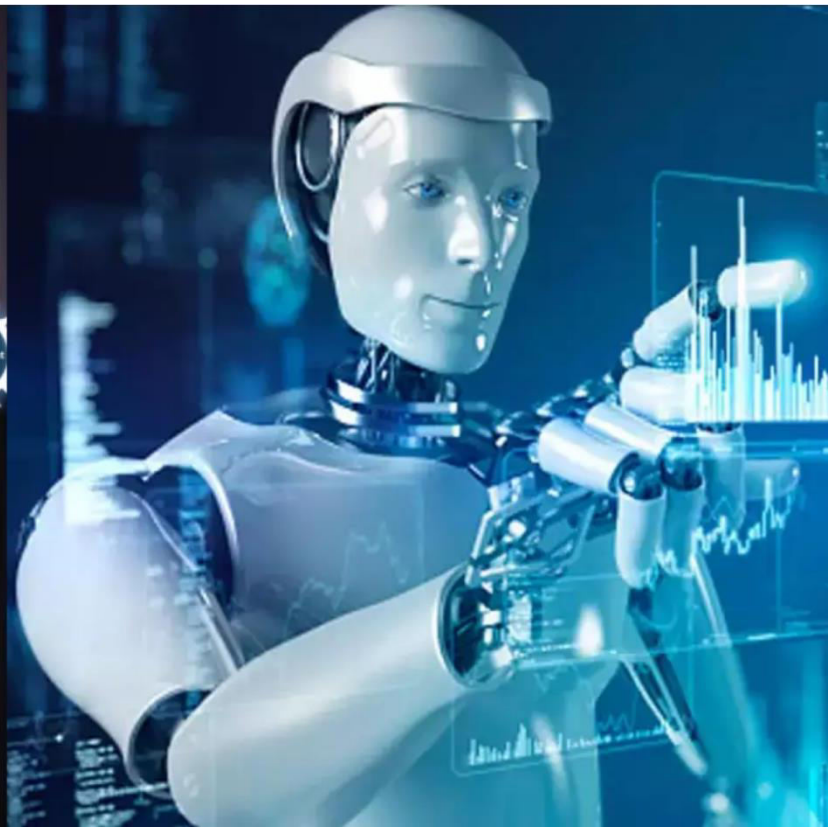
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Statistical Parameters as Probes of Phase Transitions in Two Cyano Schiff Base Liquid Crystals

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ABSTRACT: Statistical image analysis is used to find the phase transition temperatures of two thermotropic liquid crystals, p-methoxy benzylidene-p′-cyano aniline (MBCA) and p-ethoxy benzylidene-p′-cyano aniline (EBCA). Textures were recorded with a polarizing optical microscope fitted with a hot stage and a camera. Four first-order statistical parameters (mean, variance, skewness and kurtosis) and four second order statistical parameters (of gray-level co-occurrence matrix parameters - contrast, correlation, energy and homogeneity) were calculated. These parameters are computed from the textures of liquid crystals recorded as a function of temperature. In both compounds, all parameters show significant changes as a function of temperatures. The significant are known as abrupt changes indicates transitions. The texture analysis samples are carried out on MATLAB platform. From, the proposed analysis phase transitions of samples are observed: crystal to nematic transition is found at about 105 °C for MBCA and about 106 °C for EBCA. The nematic to isotropic transition, marked by a sharp peak of skewness and kurtosis followed by a uniform image, is found at about 117 °C for MBCA and about 123 °C for EBCA. The nematic range is about 11 °C for MBCA and about 17 °C for EBCA. The results show that texture statistics are a simple and sensitive way to detect phase transitions of liquid crystals.

KEYWORDS: liquid crystal, phase transition, image analysis, statistical parameters, GLCM, MBCA, EBCA

I. INTRODUCTION

Liquid crystals exhibit temperature-dependent phase transitions that are associated with changes in molecular ordering and optical textures. Accurate identification of these transitions is essential for understanding their thermal and optical behaviour. Phase transition temperatures of liquid crystals are usually measured by differential scanning calorimetry (DSC), differential thermal analysis (DTA) and polarizing optical microscopy (POM). DSC and DTA detect the heat change at a transition, so transitions with a very small enthalpy change can be missed. POM is the standard tool for identifying liquid crystal phases [1], but it is a qualitative analysis. Here, image analysis is proposed as a quantitative approach. In this analysis, texture image is converted into numbers, and the change of these numbers with temperature shows the transition. Montrucchio et al. and Eccher et al. showed that image processing can detect smooth transitions that are hard to see by eye [2,3]. Sastry and co-workers used the mean, variance, skewness and kurtosis of POM textures to find transition temperatures of several liquid crystals as well as determine the pitch in CLCs [4-7].

MBCA and EBCA are Schiff-base liquid crystals containing a cyano end group. They differ only in the length of the alkoxy end group, with methoxy and ethoxy groups, respectively. In this work, the optical textures of pure MBCA and EBCA are recorded during heating and analysed using first-order statistical parameters and second-order grey-level co-occurrence matrix (GLCM) texture parameters. First-order statistics, including mean intensity, variance, skewness, and kurtosis, describe the overall intensity distribution of the texture images. In contrast, second-order GLCM parameters, such as contrast, correlation, energy, and homogeneity, characterize spatial relationships between pixel intensities and provide information about texture uniformity, local variations, and spatial organization. Statistical image analysis is performed using the MATLAB platform, and the temperature-dependent evolution of these parameters is investigated to identify the crystal-to-nematic (Cr–N) and nematic-to-isotropic (N–I) transition temperatures of both compounds. The results are compared to evaluate how the change in alkoxy chain length influences their optical texture evolution and the associated phase transitions.

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II. EXPERIMENTAL

2.1 Materials

MBCA (p-methoxy benzylidene-p′-cyano aniline, C15H12N2O) and EBCA (p-ethoxy benzylidene-p′-cyanoaniline, C16H14N2O) were purchased from Kodak Chemical Company, USA, and used as received.

2.2 Texture recording

The textures were observed with a Meopta polarizing optical microscope fitted with a hot stage [1] and a digital camera [4]. Glass slides (75 mm × 25 mm, 1.45 mm thick) were cleaned with acetone and dried at 40 °C. Each slide was rubbed in one direction to give a unidirectional alignment. A small amount of sample was placed on the slide and heated until it became isotropic. A cover slip (0.16 mm thick) was then placed on top, and the sample was cooled to form a uniform film. The slide was placed in the hot stage between crossed polarizers and heated at 1 °C/min. Textures were recorded from 30 to 127 °C for EBCA and from 47 to 120 °C for MBCA. Images were taken every **2 °C**, with smaller steps (0.2–1 °C) near the transitions.

2.3 Image analysis

The textures were recorded as 24-bit colour images with a resolution of 1936 × 1288 pixels. The intensity of each colour band (red, green and blue) ranges from 0 to 255. Each image was converted to a gray-scale image with the intensity scaled between 0 and 1, and all parameters were calculated with MATLAB.

Let I(i, j) be the gray-level intensity of the pixel in row i and column j of an image with m rows and n columns, so the total number of pixels is $N = m \times n$ [14,15]. The following first-order statistics were calculated:

- Mean: $\mu = (1/N) \Sigma I(i, j)$
- Variance: $Var = \Sigma (I(i, j) - \mu)^2 / (N - 1)$
- Skewness: $\Sigma (I(i, j) - \mu)^3 / [(N - 1) S^3]$
- Kurtosis: $\Sigma (I(i, j) - \mu)^4 / [(N - 1) S^4]$

Here S is the standard deviation of the image. The mean is the average brightness. The variance shows how much the intensity varies around the mean. The skewness shows how symmetric the intensity distribution is, and the kurtosis shows how peaked it is (a normal distribution has a kurtosis of 3).

Four GLCM parameters were also calculated [16] contrast (the intensity difference between neighbouring pixels), correlation (the linear dependence between neighbouring pixels), energy (the uniformity of the texture) and homogeneity (how close the pixel pairs are to the diagonal of the matrix). The standard deviation was also calculated, but it is not shown because it is the square root of the variance.

III. RESULTS AND DISCUSSION

For both compounds, the mean, variance, skewness, kurtosis, contrast, correlation, energy and homogeneity of the textures were calculated at each temperature during heating. Figs 1 to 4 show each parameter for EBCA (a) and MBCA (b) side by side. The dotted lines mark the Cr–N and N–I temperatures assigned from these curves in Section 3.4. The labels Cr–N and N–I used in Sections 3.1 to 3.3 refer to those assignments.

3.1 First-order statistics

3.1.1 Mean and variance

For EBCA, the mean intensity stays between 0.14 and 0.19 and the variance between 0.004 and 0.009 from 30 to 100 °C, after which a large jump occurs at 105–106 °C—with the mean rising from 0.18 to 0.33 and reaching 0.49 at 108 °C, while the variance peaks at 0.030 at 106.8 °C—marking the Cr–N transition. Throughout EBCA's nematic range, the mean remains near 0.49–0.50, but above 117 °C it falls rapidly to 0.05 at 124 °C while the variance drops to near zero as the sample becomes isotropic. In contrast, for MBCA, the mean decreases slowly from 0.39 to 0.34 and the variance from 0.014 to 0.011 between 47 and 103 °C, before the mean drops sharply between 104 and 106 °C (0.34, 0.27, 0.16, and 0.11 at 104, 105, 105.3, and 106 °C) to signify its Cr–N transition. From 106 to 111 °C, MBCA's mean remains around 0.09 while its variance decreases from 0.0079 to 0.0021, followed by a secondary rise in mean to 0.37 at 112.5–113 °C and a variance maximum of 0.039 at 112.7 °C. Finally, both parameters for MBCA decrease, reaching

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a mean of 0.066 and a variance of 1.5×10^{-4} at 117 °C, and settling at 0.055 and 4.5×10^{-5} at 120 °C, representing a uniform dark image.

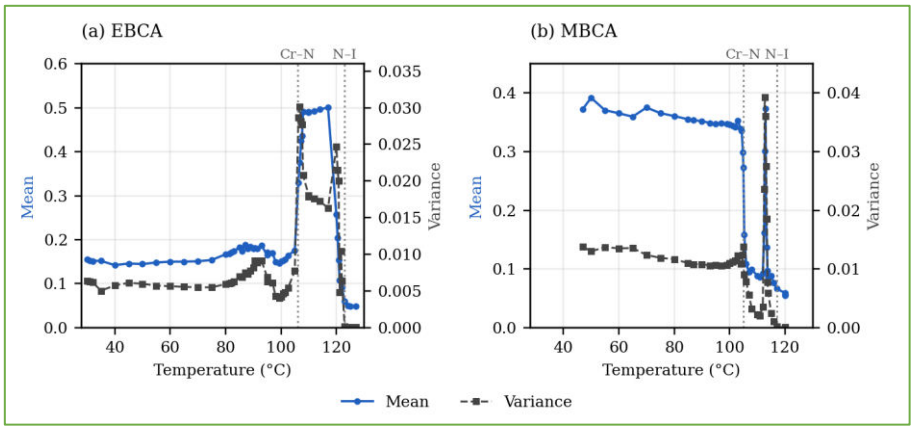


Fig.1. Mean and variance as a function of temperature for (a) EBCA and (b) MBCA. Dotted lines mark the Cr–N and N–I temperatures.

3.1.2 Skewness and kurtosis

In the crystalline phase of EBCA, the skewness ranges between 1 and 1.7, while the kurtosis steadily increases from about 3.9 to 6.9 up to 105 °C. At the Cr–N transition, the kurtosis drops to 2.0 and the skewness to +0.6, eventually shifting to negative values (−0.7) within the nematic range. Upon reaching the N–I transition at 122 °C, both parameters exhibit a sharp peak, with skewness reaching 4.0 and kurtosis peaking at 20.6, before returning to post-clearing baseline values of approximately −0.3 and 3.3, respectively, corresponding to a uniform dark image. For MBCA, both parameters rise slowly in the crystalline phase (skewness from 0.31 to 0.59 and kurtosis from 2.4 to 2.8 between 47 and 103 °C), before stepping up to 0.76 and 3.2 at 104 °C and climbing steeply through the Cr–N transition. Across MBCA's dark nematic range, they continue rising to reach 5.6 and 42.8 at 111 °C as the texture becomes nearly black with only sparse bright pixels. They then temporarily drop to −0.31 and 1.6 at 113 °C where the texture brightens, before sharply escalating to mark the N–I clearing at 117 °C with an intense peak (skewness of 17.2 and kurtosis of 599). Beyond clearing at 120°C, MBCA's skewness settles between −0.7 and −0.3 with a kurtosis around 3, mirroring EBCA's isotropic state; because MBCA's kurtosis spans over two orders of magnitude, it is presented on a logarithmic scale in Fig 2.

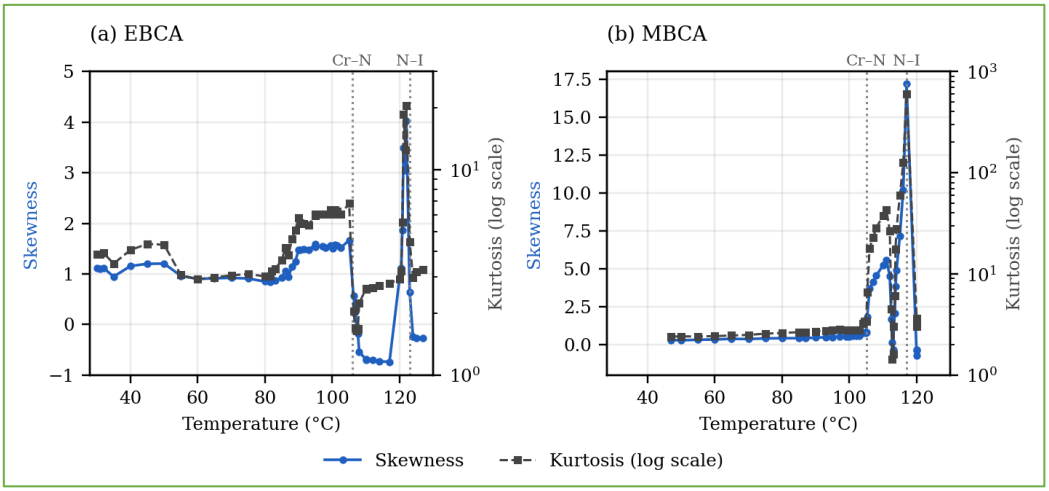


Fig.2. Skewness and kurtosis (log scale) as a function of temperature for (a) EBCA and (b) MBCA Dotted lines mark the Cr–N and N–I temperatures.

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3.2 GLCM (second-order) texture parameters

3.2.1 Energy and homogeneity

In EBCA, energy and homogeneity are about 0.2 and 0.8 in the crystalline phase, fall to 0.10–0.15 and 0.80–0.82 in the nematic phase, and rise steeply above 120 °C to reach 1.0 at 124 °C. In MBCA, they are about 0.12–0.16 and 0.77–0.79 up to 104 °C. At the Cr–N transition they rise sharply (energy 0.15, 0.31 and 0.65, and homogeneity 0.79, 0.90 and 0.95 at 105, 105.3 and 106 °C) and reach 0.85 and 0.98 at 111 °C. They dip to 0.18 and 0.92 at 112.7 °C, where the bright texture appears, and then rise to 0.998 and 0.9997 at 117 °C and to 1 at 120 °C (Fig 3).

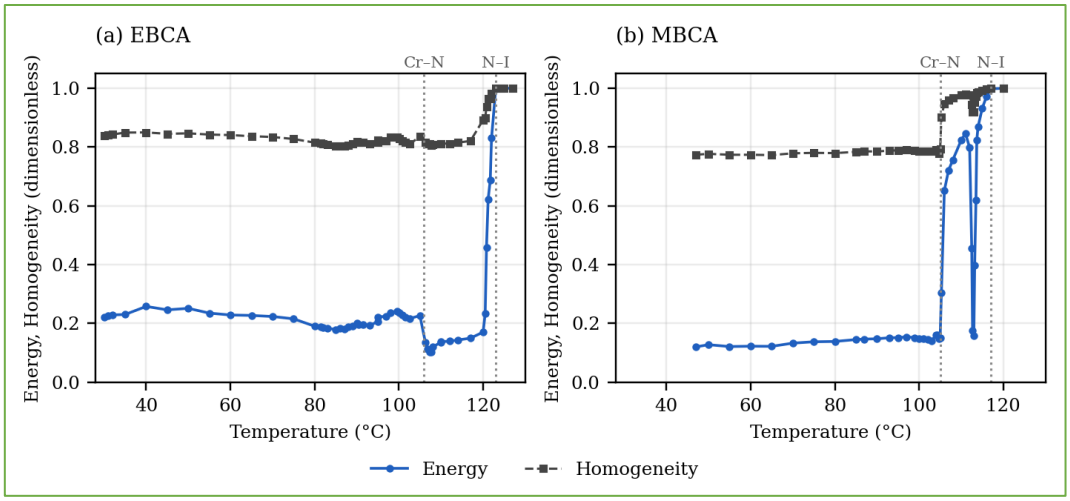


Fig.3. Energy and homogeneity as a function of temperature for (a) EBCA and (b) MBCA Dotted lines mark the Cr–N and N–I temperatures.

3.2.2 Contrast and correlation

In EBCA, the correlation jumps from 0.68 to 0.87 at the Cr–N transition, and the contrast falls from 0.43 at 117 °C to 0.24 at 120 °C and to zero at 124 °C. In MBCA, the contrast is 0.5–0.6 in the crystalline phase with a maximum of 0.62 at 104.8 °C, then drops to 0.24 at 105.3 °C and to 0.05 at 110 °C. It rises briefly to 0.17–0.18 at 112.7–113 °C and falls to 0.0013 at 117 °C and to 0 at 120 °C. The MBCA correlation rises from 0.66–0.70 to 0.88 at 106 °C and to a maximum of 0.98 at 113.5 °C, and falls to 0.87 at 117 °C. Above the clearing point the contrast is zero, so the correlation is not defined and is not plotted (Fig 4).

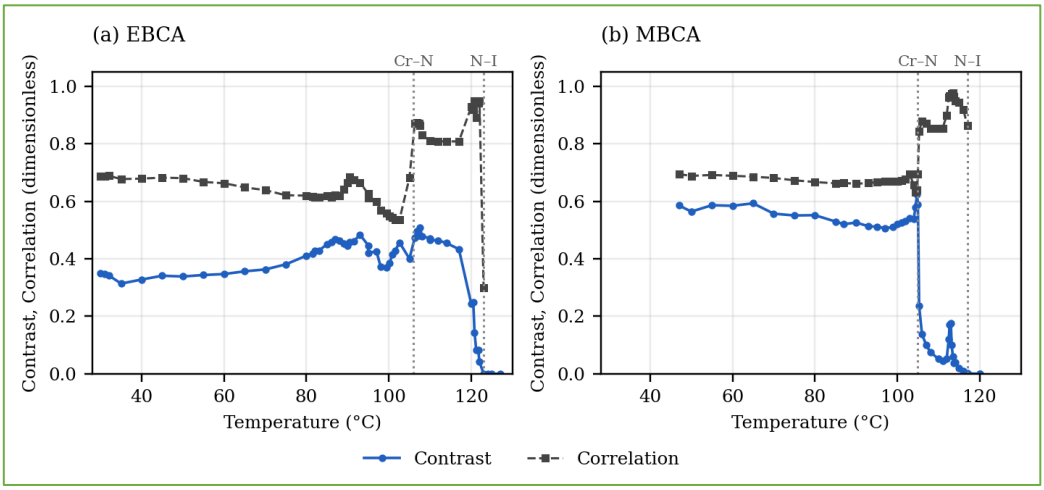


Fig 4. Contrast and correlation as a function of temperature for (a) EBCA and (b) MBCA. Dotted lines mark the Cr–N and N–I temperatures. The correlation is undefined when the contrast is zero.

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3.4 Transition temperatures

The parameters discussed in Sections 3.1 to 3.3 change abruptly at the same two temperatures in both compounds, so the assignment does not depend on one parameter. The transition temperatures are given in Table 1. For both compounds, the N–I transition is spread over several degrees, from the first change in the texture (onset) to the point where the image is almost uniform (clearing). The clearing temperature, taken as the first temperature at which the contrast is below 0.002 and the energy is above 0.99, is used as the N–I temperature.

Table.1. Phase transition temperatures of EBCA and MBCA from image analysis on heating.

Compound	Cr–N (°C)	N–I onset (°C)	N–I clearing (°C)	Nematic range (°C)
EBCA	106 (105–106.3)	120	123	17
MBCA	105 (104–106)	112	117	11

The same transition looks different in the two compounds. At the Cr–N transition the mean of EBCA rises from 0.18 to 0.49, but the mean of MBCA falls from 0.35 to 0.11 and the image becomes almost uniform. The brightness of a texture between crossed polarizers depends on how the director is aligned in the cell. A nematic film aligned along a polarizer axis, or perpendicular to the glass plates, looks dark. The direction of the change in the mean therefore does not identify the phase, and only the abrupt change itself is used here.

The MBCA image between 106 and 112 °C is assigned to the nematic phase, not to the isotropic liquid, for two reasons. First, it is not completely uniform (contrast 0.05–0.14, energy 0.65–0.85). Second, the bright texture returns at 112.5–113.5 °C, which cannot happen on heating from the isotropic liquid. In the isotropic state the contrast is exactly 0 and the energy is 1, which is reached at 120 °C. At 117 °C the image is already almost uniform (contrast 0.0013, energy 0.998). This agrees with the X-ray result that the structure of MBCA changes sharply only at the N–I transition [6].

The two compounds behave in a similar way. Both show a sharp Cr–N change at almost the same temperature (about 106 °C for EBCA and 105 °C for MBCA). Both also end with a sharp peak of skewness and kurtosis, followed by a uniform image (contrast 0, energy 1, homogeneity 1). The main difference is in the nematic phase. The N–I temperature of MBCA (about 117 °C) is about 6 °C lower than that of EBCA (about 123 °C), so the nematic range is shorter (about 11 °C against about 17 °C). The N–I transition of MBCA also starts earlier, at 112 °C against 120 °C for EBCA. MBCA shows a short bright event at 112.5–113.5 °C before clearing, which EBCA does not show. It may be a change in the alignment of the film. EBCA also shows weak changes near 55, 80 and 97 °C. They are much smaller than the two main transitions, so they are not assigned as phase transitions.

For EBCA, a previously reported study identified a nematic phase over the temperature range of 74–112.5 °C [11]. In the present study, the N–I transition temperature is approximately 123 °C, which is about 10 °C higher than the reported value, while the Cr–N transition temperature of approximately 106 °C is substantially higher than the reported lower limit of 74 °C. These discrepancies may arise from differences in experimental conditions, including temperature calibration, heating rate (1 °C/min in the present study), sample thickness, surface conditions, and sample purity. Notably, the incorporation of ZnO nanoparticles has been reported to increase the N–I transition temperature of EBCA demonstrating that material composition can influence its thermal behaviour. A weak change in the image-statistical parameters near 80 °C is also observed in the present data; however, its magnitude is considerably smaller than the pronounced change near 106 °C. Therefore, the feature near 80 °C cannot be conclusively assigned to the Cr–N transition based on the image-statistical results alone. Further investigation using complementary techniques would be necessary to clarify the origin of this feature and establish the transition temperatures more reliably.

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IV. CONCLUSION

In this study, first-order statistical parameters (mean intensity, variance, skewness, and kurtosis) and second-order GLCM texture parameters (contrast, correlation, energy, and homogeneity) were extracted from POM images of MBCA and EBCA during heating using MATLAB-based image analysis. The temperature-dependent variations in these parameters revealed two distinct transition regions corresponding to the crystal-to-nematic (Cr–N) and nematic-to-isotropic (N–I) transitions. The N–I transition was particularly evident from the sharp changes in skewness and kurtosis, accompanied by the disappearance of characteristic birefringent textures as the samples entered the isotropic phase. The nearly identical Cr–N transition temperatures but different N–I transition temperatures indicate that the two compounds exhibit distinct thermal ranges of nematic ordering despite their similar initial transition behaviour. These differences may be associated with the change in alkoxy chain length, although further measurements are required to establish the underlying molecular mechanisms. Overall, the results demonstrate that statistical analysis of POM images provides a quantitative and objective approach for identifying phase-transition temperatures in liquid crystals. Furthermore, the higher absolute transition temperatures reported for EBCA compared with some literature values and are in good agreement. Therefore, the present findings primarily establish the potential of first- and second-order image statistics for distinguishing phase transitions and comparing the thermal behaviour of structurally related liquid crystals.

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