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Phenomenological Theory of Isotropic-Genesis Nematic Elastomers

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We consider the impact of the elastomer network on the nematic structure and fluctuations in isotropic-genesis nematic elastomers, via a phenomenological model that underscores the role of network compliance. The model contains a network-mediated nonlocal interaction as well as a new kind of random field that reflects the memory of the nematic order present at cross-linking, and also encodes local anisotropy due to localized polymers. This model enables us to predict a regime of short-ranged oscillatory spatial correlations (both thermal and glassy) in the nematic alignment.

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Consider a melt or solution of nematogenic polymers, by which we mean long, flexible polymers carrying rod-like units. These units, which give the system the possibility of exhibiting liquid crystallinity, may be integrated along the polymer chain backbones (the main-chain case) or in groups that dangle from the backbone (the side-chain or pendant case). Now consider the process of instantaneous cross-linking. Here, one begins with the melt or solution at equilibrium and—so rapidly that hardly any relaxation has time to occur—one introduces permanent bonds between some random fraction of the pairs of chain segments that happen, at the instant of cross-linking, to be nearby one another. When the cross-linking process is carried out in the *isotropic* state of the nematogens, the resulting material is called an *isotropic-genesis nematic elastomer* (or IGNE; see Refs. [1–3]). The IGNE is a macroscopic random network medium that "memorizes" both the positions of the chain segments and the orientations of the nematogen units at the instant of cross-linking. This memorization is, however, only partial, as a result of the thermal fluctuations that occur in the new, post-cross-linking equilibrium state.

Several prior approaches to the liquid crystallinity of such materials [4–7] have assumed the presence of a random field that is attached to an elastically deformable medium, but they ignore the following two facts: (i) the medium is itself liquid-like at short lengthscales, owing to thermal position fluctuations of the network chain segments; and (ii) the random field is influenced by the configuration of nematic alignments at the instant of cross-linking, owing to the afore-mentioned memory effect. In this paper, we focus on the influence of such short-lengthscale liquidity and memorization on liquid crystallinity in IGNEs. We construct a suitable Landau-type free energy [see Eq. (3)], which involves two novel elements: (i) a lengthscale-dependent nematic-nematic term reflecting the short-lengthscale liquidity; and (ii) a random field that takes the memory effect into account. We show that these elements lead to three predictions: (i) the correlation length of the thermal nematic fluctuations in an IGNE having a weak random field is *shorter* than it is in *liquid* nematics held at the same temperature; (ii) the thermal and glassy correlations of the liquid crystallinity in IGNEs having sufficiently strong random fields exhibit *oscillatory* spatial decay; and (iii) when the local nematic order present at the instant of cross-linking is spatially correlated over distances larger than the typical localization length of the network, the system strongly memorizes that local nematic order. We expect these features all to be detectable via light scattering experiments

To describe the structure and correlations of the system post cross-linking, we employ the local nematic order parameter $Q_{dd'}(\mathbf{r})$, which is traceless, symmetric, and of rank-two, and is defined microscopically via

$$Q_{dd'}(\mathbf{r}) = \sum_{p=1}^{P} \left(N_d^p N_{d'}^p - D^{-1}\delta_{dd'} \right) \delta^{(D)}\big(\mathbf{r} - \mathbf{R}^p\big), \quad (1)$$

where P is the number of rod-like units, $\mathbf{N}^p$ is the microscopic unit orientation vector of unit p and $\mathbf{R}^p$ is its microscopic position vector in D dimensions, and $\mathbf{r}$ is an arbitrary position vector. In addition, we characterize the random local environmental anisotropy, which tends to induce local nematic alignment $\mathbf{Q}$ in the post cross-linking system, in terms of the random tensor field $\mathbf{M}$:

$$\mathbf{M}(\mathbf{r}) = \mathbf{Y}(\mathbf{r}) + \frac{T}{T_p} \int d^D r' \, \mathrm{H}(\mathbf{r} - \mathbf{r}') \, \mathbf{Q}^0(\mathbf{r}'). \quad (2)$$

Here, T is the measurement temperature (i.e., the temperature at which the system is maintained, in equilibrium, long after the cross-linking process), and T_p is the temperature of the equilibrium state into which cross-links are instantaneously created, where p stands for preparation. The random environmental anisotropy described by $\mathbf{M}$ is caused by the thermally averaged part of random local spatial arrangement of the localized polymers at post-cross-linking equilibrium. It consists of two parts: (i) a part that is independent of the the pattern of local nematic alignment $\mathbf{Q}^0$ present at

the instant of cross-linking, which we call the *memory-independent random field* and denote by $\mathbf{Y}$; and (ii) a part that is due to the pattern of $\mathbf{Q}^0$, which we call the *memory-dependent random field*. $\mathbf{Q}^0$ is partially imprinted in the network structure, and this imprint then partially elicits a response similar to $\mathbf{Q}^0$ in the post-cross-linking state. The relationship between $\mathbf{Q}$ and $\mathbf{Q}^0$ is characterized by a "smearing" kernel, which embodies the idea that $\mathbf{Q}$ (i.e., the post-cross-linking equilibrium-state memory of $\mathbf{Q}^0$) is partially erased, as a result of the thermal position fluctuations of the network. Equivalently, viewed from wave-vector space, the contribution from $\mathbf{Q}^0$ becomes $\mathrm{H}_{\mathbf{k}}\mathbf{Q}^0_{\mathbf{k}}$. Physically, we expect $\mathrm{H}(\boldsymbol{r})$ to be positive and bell-shaped, operative primarily over a region of order the typical localization length ξ_L (which reflects how weakly localized the network constituents are; see, e.g., Ref. [8]), and to decay monotonically with increasing $|\boldsymbol{r}|$ over this lengthscale, ultimately tending to zero for $|\boldsymbol{r}| \gg \xi_L$. Correspondingly, in wave-vector space $\mathrm{H}_{\mathbf{k}}$ would decay monotonically to zero over a scale ξ_L^{-1}. Hence, we see that H serves as a "soft filter," deamplifying—more strongly the shorter the lengthscale—the contributions made by the Fourier components of $\mathbf{Q}^0$ to the random anisotropic environment on distance scales shorter than ξ_L. This is a natural consequence of the liquid-like character of the post-cross-linking system on lengthscales shorter than ξ_L. As for the overall *amplitude* of H, this we expect to *increase* with (i) the fraction G of polymers that are localized; (ii) the sharpness of localization, $1/\xi_L$; (iii) the nematogen-nematogen aligning interaction J; and (iv) the length ℓ of the nematogens; and we expect this amplitude to *decrease* with the "measurement temperature" T (see below for more on this concept), because thermal fluctuations tend to moderate any aligning forces. A complementary microscopic calculation [9] bears out these expectations, yielding $\mathrm{H}_{\mathbf{k}} = \mathrm{H}_{\mathbf{0}} \exp(-k^2\xi_L^2/2)$, where the amplitude $\mathrm{H}_{\mathbf{0}} \propto G^2J^2(\ell/\xi_L)^4/T$.

In terms of these ingredients, we take as a model for the Landau-type free-energy cost F associated with the induction of local nematic order in the post-cross-linking system the form:

$$F = \frac{1}{2}\int_{\mathbf{k}} \Big(\big(\mathcal{A}t + \mathcal{L}k^2 + \mathrm{H}_{\mathbf{k}}\big)\big\{\mathbf{Q}_{\mathbf{k}}\mathbf{Q}_{-\mathbf{k}}\big\} - 2\big\{\big(\mathbf{Y}_{\mathbf{k}} + (T/T_p)\,\mathrm{H}_{\mathbf{k}}\,\mathbf{Q}^0_{\mathbf{k}}\big)\mathbf{Q}_{-\mathbf{k}}\big\}\Big). \quad (3)$$

Here, $\int_{\mathbf{k}}$ is shorthand for $\int d^Dk/(2\pi)^D$, k^2 is the squared length of the vector $\mathbf{k}$, and the $R_{\mathbf{k}}$ is the Fourier transform $\int d^Dr\, R(\mathbf{r})\, \exp(i\mathbf{k}\cdot\mathbf{r})$. In addition, curly brackets—as in $\{\mathbf{S}\,\mathbf{S}'\}$—indicate the trace of the product of the tensors $\mathbf{S}$ and $\mathbf{S}'$, i.e., $\sum_{d,d'=1}^{D} S_{dd'}S'_{d'd}$. Furthermore, $\mathcal{A}$ characterizes the aligning tendencies of nematic freedoms; and $\mathcal{L}$ is the generalized stiffness for nematic order, for which (for the sake of simplicity) we have adopted the Landau-de Gennes equivalent of the one-Frank-constant approximation [10]. The symbol t denotes the reduced measurement temperature [11]; the occurrence of two temperatures, T and T_p, stems from the fact that elastomers and related systems are characterized by not one but two statistical ensembles. One, which we call the *preparation ensemble*, provides a statistical description of the random (non-equilibrating, unmeasured) freedoms $\mathbf{Q}^0$ that characterize the local alignment immediately prior to cross-linking. The other ensemble describes the equilibrium state of the system long after cross-linking was done, via the statistics of the equilibrating variables $\mathbf{Q}$; we call it the *measurement ensemble*.

The free energy (3) consists of two terms. The first two elements of the first term constitute the familiar Landau-de Gennes free energy at quadratic order; higher-order terms have been neglected as we focus on the properties of IGNEs at $t > 0$. These elements describe the free-energy cost of inducing nematic alignment from the unaligned state. The second term incorporates what we have described above, viz., the influences of (i) the configuration of the rod-like constituents at the instant of cross-linking, via $\mathbf{Q}^0$, together with (ii) the memory-independent random field $\mathbf{Y}$ caused by the localized polymers post cross-linking. From the (previously given) value of $\mathrm{H}_{\mathbf{0}}$ and Eq. (3), we see that the contribution to F/T involving $\mathbf{Q}^0$ carries a factor $(J/T_p)(G\ell^2/\xi_L^2)^2(J/T)$. In it, the two temperature factors show that the network is better able to store a given pattern $\mathbf{Q}^0$ the lower the preparation temperature T_p and, similarly, better able to elicit $\mathbf{Q}^0$ from $\mathbf{Q}$ the lower the measurement temperature T. Taking the two terms together, F is minimized by the most probable nematic configuration $\widetilde{\mathbf{Q}}$, which is given by

$$\widetilde{\mathbf{Q}}_{\mathbf{k}} = \big(\mathbf{Y}_{\mathbf{k}} + (T/T_p)\,\mathrm{H}_{\mathbf{k}}\,\mathbf{Q}^0_{\mathbf{k}}\big)/\big(\mathcal{A}t + \mathcal{L}k^2 + \mathrm{H}_{\mathbf{k}}\big). \quad (4)$$

By completing the square with respect to the first and second terms in Eq. (3), we arrive at the following form for F (up to a non-thermally fluctuating term):

$$\frac{1}{2}\int_{\mathbf{k}} \big(\mathcal{A}t + \mathcal{L}k^2 + \mathrm{H}_{\mathbf{k}}\big)\big\{\big(\mathbf{Q}_{\mathbf{k}} - \widetilde{\mathbf{Q}}_{\mathbf{k}}\big)\big(\mathbf{Q}_{-\mathbf{k}} - \widetilde{\mathbf{Q}}_{-\mathbf{k}}\big)\big\}. \quad (5)$$

The third element in the first term of the free energy (3) is a new and central element. It encodes the essential physical difference between our model and previous models of IGNEs, viz., the elastomer's possession of a network that is localized randomly and fluctuating thermally, and is, furthermore, *liquid-like at sub-localization-length scales* and solid-like at larger scales. As can be seen from Eq. (5), this element gives rise to a nonlocal free-energy cost for creating a departure from the nematic pattern $\widetilde{\mathbf{Q}}_{\mathbf{k}}$. This cost arises because the network mediates additional nematic-nematic interactions. We emphasize that: (i) the mediated interactions addressed here are not of a type transmitted through coupling between the nematic order and elastic deformation, but of a novel type that

is related to the short-lengthscale liquidity feature of the network [12]; and (ii) the free-energy cost associated with such mediated interactions arises from the competition of the tendency for nematic alignment with localization forces (associated with short-range liquidity) rather than with elastic forces [13]. Thus, the nonlocal energy cost of creating a nematic departure from $\widetilde{\mathbf{Q}}$ that is essentially uniform over a lengthscale rather larger than ξ_L is relatively large, as at this lengthscale the solidness of the network becomes pronounced. Conversely, the nonlocal energy cost is relatively mild if the departure varies only over some lengthscale rather shorter than ξ_L, where the system has a more liquid-like character.

Various averaged diagnostics of the system involving the local nematic order $\mathbf{Q}(\mathbf{r})$ may be considered by means of F. These averages come in two types. First, there are disorder-averaged quantities (denoted by $[\cdots]$), by which we mean quantities averaged over suitably distributed $\mathbf{Y}$ and $\mathbf{Q}^0$. Second, there are thermally averaged quantities (denoted by $\langle\cdots\rangle$), by which we mean quantities averaged over the measurement ensemble. We focus on two particular diagnostics of nematic elastomers. The first is the thermal fluctuation correlator $\mathcal{C}^T$, defined [14] via

$$\mathcal{C}^T(\boldsymbol{r}-\boldsymbol{r}') \equiv \big[\big\langle\big\{\big(\mathbf{Q}(\boldsymbol{r})-\langle\mathbf{Q}(\boldsymbol{r})\rangle\big)\big(\mathbf{Q}(\boldsymbol{r}')-\langle\mathbf{Q}(\boldsymbol{r}')\rangle\big)\big\}\big\rangle\big]. \tag{6a}$$

The second is the glassy correlator $\mathcal{C}^G$, defined via

$$\mathcal{C}^G(\boldsymbol{r}-\boldsymbol{r}') \equiv \big[\big\{\big\langle\mathbf{Q}(\boldsymbol{r})\big\rangle\big\langle\mathbf{Q}(\boldsymbol{r}')\big\rangle\big\}\big], \tag{6b}$$

which is a diagnostic of any randomly frozen (i.e., time-persistent) nematic order present. In particular, the value $\mathcal{C}^G(\boldsymbol{r})|_{\boldsymbol{r}=\mathbf{0}}$ is the nematic analog of the Edwards-Anderson order parameter for spin glasses [15], and measures the magnitude of the local frozen nematic order; hence the name *glassy correlator*. Moreover, how $\mathcal{C}^G(\boldsymbol{r})$ varies with $\boldsymbol{r}$ determines the spatial extent of regions that share a roughly common nematic alignment [16].

One could also consider the disorder-averaged quantity $[\langle\mathbf{Q}(\boldsymbol{r})\rangle]$; it, however, vanishes, owing to the macroscopic isotropy of the post-cross-linking state. On the other hand, the thermal average of the local order parameter for a specific realization of the quenched disorder $\langle\mathbf{Q}(\boldsymbol{r})\rangle$ is maintained at a nonzero, time-persistent, random value, which we shall compute shortly. This nonzero value is the result of the partial trapping, by the network, of the orientational randomness $\mathbf{Q}^0$ present at the instant of cross-linking, together with the memory-independent random field $\mathbf{Y}$ of the network, post cross-linking. The free energy (3) is quadratic in $\mathbf{Q}$, and therefore the computation of $\langle\mathbf{Q}\rangle$ and $\mathcal{C}^T$ using the weight $\exp(-F/T)$ is elementary, yielding $\langle\mathbf{Q}_{\mathbf{k}}\rangle = \widetilde{\mathbf{Q}}_{\mathbf{k}}$ and

$$\big\langle\big\{\big(\mathbf{Q}_{\mathbf{k}}-\langle\mathbf{Q}_{\mathbf{k}}\rangle\big)\big(\mathbf{Q}_{\mathbf{k}'}-\langle\mathbf{Q}_{\mathbf{k}'}\rangle\big)\big\}\big\rangle = \frac{T\mu_D\,\delta_{\mathbf{k}+\mathbf{k}',\mathbf{0}}}{\mathcal{A}t+\mathcal{L}k^2+\mathrm{H}_{\mathbf{k}}}. \tag{7}$$

Here, $\mu_D \equiv (D-1)(D+2)/2$ counts the number of degrees of freedom of $\mathbf{Q}$ and takes the value 5 for $D=3$. Note that we have chosen units in which Boltzmann's constant has the value unity.

To perform the average over the quenched random variables $\mathbf{Y}$ and $\mathbf{Q}^0$ we must adopt a model for their statistics that is consistent with the physical origin each has. The choice we make is that $\mathbf{Y}$ and $\mathbf{Q}^0$ are independent, Gaussian-distributed random fields, with zero means and non-zero variances, the latter being given by

$$\big[\big\{\mathbf{Q}^0_{\mathbf{k}}\,\mathbf{Q}^0_{\mathbf{k}'}\big\}\big] = T_p\mu_D\,\frac{\delta_{\mathbf{k}+\mathbf{k}',\mathbf{0}}}{\mathcal{A}^0t_p+\mathcal{L}^0k^2}, \tag{8a}$$

$$\big[\big\{\mathbf{Y}_{\mathbf{k}}\,\mathbf{Y}_{\mathbf{k}'}\big\}\big] = T\,\mathrm{H}_{\mathbf{k}}\,\delta_{\mathbf{k}+\mathbf{k}',\mathbf{0}}\,. \tag{8b}$$

Here, $\mathcal{A}^0$ and $\mathcal{L}^0$ are, respectively, the preparation-ensemble counterparts to $\mathcal{A}$ and $\mathcal{L}$. The statistics of $\mathbf{Q}^0$ depends on the reduced temperature of the preparation ensemble, t_p [11]; it does not depend on H, because H encodes the physics of random but imperfect spatial localization, and this only comes into being post cross-linking. (The *impact* of $\mathbf{Q}^0$ *does* depend on H, as H controls the relaxation of $\mathbf{Q}$ from $\mathbf{Q}^0$ to its equilibrium value, post cross-linking.) By contrast, the statistics of $\mathbf{Y}$ *does* depend on H; this is because H characterizes the typical value of the memory-independent random field that results from the random (imperfect) spatial localization of the polymers constituting the network. In view of their distinct origins it is natural that $\mathbf{Y}$ and $\mathbf{Q}^0$ be statistically uncorrelated. However, it is not a coincidence (and can indeed be derived from a microscopic calculation [9]) that the H that characterizes the *orientational caging* induced by the network (via $\mathbf{Y}$) is the same H that determines the fidelity with which the network preserves the *orientational order* present immediately post cross-linking (i.e., $\mathbf{Q}^0$). It is, in fact, natural, because localization that is sharper and more widespread (i.e., involves a larger localized fraction) both *creates* more intense network-induced orientational caging and *enhances* the trapping-in of the local nematic order present immediately post cross-linking. Our physical expectation, borne out by a complementary microscopic analysis (see Ref. [9]), is that such strengthening of the localization would enhance memorization more strongly than it would orientational caging. This expectation is consistent with the phenomenological choice presented here, in which the corresponding contributions to the random anisotropy field, Eq. (2), scale as $\sqrt{\mathrm{H}}$ for the caging (i.e., $\mathbf{Y}$) part and H for the "memorization" (i.e., $\mathbf{Q}^0$) part.

Returning to the disorder-averaged diagnostics—the mean $[\langle\mathbf{Q}\rangle]$ and the correlators $\mathcal{C}^T$ and $\mathcal{C}^G$—we complete their computation using the statistics of the quenched disorder, Eqs. (8), to arrive at (with $[\langle\mathbf{Q}_{\mathbf{k}}\rangle]=0$)

$$\mathcal{C}^T_{\mathbf{k}} = T\mu_D\,\frac{1}{\mathcal{A}t+\mathcal{L}k^2+\mathrm{H}_{\mathbf{k}}}, \tag{9a}$$

$$\mathcal{C}^G_{\mathbf{k}} = T\mu_D\,\frac{\frac{T}{T_p}(\mathcal{A}^0t_p+\mathcal{L}^0k^2)^{-1}|\mathrm{H}_{\mathbf{k}}|^2+\mathrm{H}_{\mathbf{k}}}{\big(\mathcal{A}t+\mathcal{L}k^2+\mathrm{H}_{\mathbf{k}}\big)^2}. \tag{9b}$$

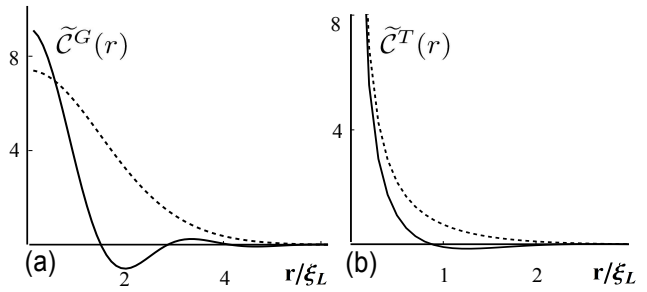

FIG. 1: (a) Glassy correlator (rescaled) $\widetilde{\mathcal{C}}^G(r) \equiv (60\pi^2\mathcal{L}/T\mu_D)\mathcal{C}^G(r)$, for $t_p \gg T\mathrm{H}_\mathbf{0}/T_p\mathcal{A}^0$; $t = 0.1\mathcal{L}/\mathcal{A}\xi_L^2$, at (i) $\mathrm{H}_\mathbf{0}/\mathrm{H}^{(c)} = 0.5$ (weak disorder; red, dashed) and (ii) $\mathrm{H}_\mathbf{0}/\mathrm{H}^{(c)} = 40$ (strong disorder; blue, solid). (b) Thermal correlator (rescaled) $\widetilde{\mathcal{C}}^T(r) \equiv (2\pi^2\mathcal{L}/T\mu_D)\mathcal{C}^T(r)$, for the same parameters. On going from weak to strong disorder, both correlators cross over from simple exponential decay to oscillatory decay at wavelength of order ξ_L.

Disorder strength	Weak ($\mathrm{H}_\mathbf{0} < \mathrm{H}^{(c)}$)	Strong ($\mathrm{H}_\mathbf{0} > \mathrm{H}^{(c)}$)
$\xi^2_{T,o}$	∞	$\frac{1}{2}\xi_L^2/\ln(\mathrm{H}_\mathbf{0}/\mathrm{H}^{(c)})$
$\xi^2_{T,d}$	$\xi_N^2\frac{1-(\mathrm{H}_\mathbf{0}/\mathrm{H}^{(c)})}{1+(\mathrm{H}_\mathbf{0}/\mathcal{A}t)}$	$\sim \xi_L^2/\left(1+\frac{\xi_L^2}{2\xi_N^2}\right)$
$\xi^2_{G,o}$	∞	$\sim \xi_L^2/\ln(\mathrm{H}_\mathbf{0}/\mathrm{H}^{(c)})$
$\xi^2_{G,d}$	$\frac{1}{2}\xi_L^2 + 2\xi_N^2\frac{1-(\mathrm{H}_\mathbf{0}/\mathrm{H}^{(c)})}{1+(\mathrm{H}_\mathbf{0}/\mathcal{A}t)}$	$\sim \xi_L^2$

TABLE I: Values of the correlation lengthscales ($\xi_{T,d}$ and $\xi_{G,d}$), and the oscillation wavelengths ($\xi_{T,o}$ and $\xi_{G,o}$), in the weak– and strong–disorder regimes for the case of IGNEs crosslinked at $t_p \gg T\mathrm{H}_\mathbf{0}/T_p\mathcal{A}^0$.

Having computed the correlators $\mathcal{C}^T$ and $\mathcal{C}^G$, we now set about using them to study how the presence of a network modifies the organizational behavior of nematic freedoms. To do this, we first note that there are two emergent lengthscales present in IGNEs: (i) the typical localization length, ξ_L, quantifying the sharpness of localization of polymers belonging to the network; and (ii) the *intrinsic nematic correlation length*, ξ_N [$\equiv \sqrt{\mathcal{L}/\mathcal{A}t}$], describing the range over which nematic freedoms would be correlated if there were no network present. On the other hand, we have the strength of the memory-independent random field $\mathbf{Y}$, which is characterized by $\sqrt{\mathrm{H}_\mathbf{0}}$. In what follows, we shall study the dependence of $\mathcal{C}^T$ and $\mathcal{C}^G$ on the parameters ξ_N, ξ_L and $\mathrm{H}_\mathbf{0}$, doing so for two specific systems: one prepared at $t_p \gg T\mathrm{H}_\mathbf{0}/T_p\mathcal{A}^0$, and one at $t_p < T\mathrm{H}_\mathbf{0}/T_p\mathcal{A}^0$.

First consider the behaviors of $\mathcal{C}^T$ and $\mathcal{C}^G$ for $t_p \gg T\mathrm{H}_\mathbf{0}/T_p\mathcal{A}^0$, so that any local nematic order present immediately post cross-linking (and thus available for trapping in) is spatially correlated only over distances far shorter than the typical localization length ξ_L; see Table I. This separation of lengthscales implies that the local nematic order arising from $\mathbf{Q}^0$ would be heavily "washed out" by thermal fluctuations of the network. Thus, in this regime the dominant contribution to the trapped-in local nematic order originates in the memory-independent random field, $\mathbf{Y}$.

Continuing with the case $t_p \gg T\mathrm{H}_\mathbf{0}/T_p\mathcal{A}^0$, we observe that $\mathcal{C}^T$ and $\mathcal{C}^G$ exhibit qualitatively distinct behaviors in two regimes, depending on the strength of the random field (see Fig. 1). For $\mathrm{H}_\mathbf{0} < \mathrm{H}^{(c)}$ (where $\mathrm{H}^{(c)} \equiv 2\mathcal{L}/\xi_L^2$—the *weak disorder regime*), $\mathcal{C}^T$ and $\mathcal{C}^G$ *decay simply* with increasing real-space separation. More specifically, by examining their small wave-vector behaviors we ascertain that the respective associated correlation lengths $\xi_{T,d}$ and $\xi_{G,d}$ have the values given in Table I. We see from the behavior of $\xi_{T,d}$ the physically reasonable result that the random network, with its thermal fluctuations, serves to shorten the nematic thermal fluctuation correlation length from the value it would have in the absence of the network, a phenomenon that conventional (i.e., non-thermally fluctuating) random-field approaches would not capture. As for $\xi^2_{G,d}$, it comprises two parts. One ($\propto \xi^2_{T,d}$) arises from the nematic thermal correlations; the other ($\propto \xi_L^2$) comes from the local aligning effect exerted by the cage. The fact that $\xi_{G,d}$ increases with ξ_L does not mean that a more weakly crosslinked network (for which ξ_L would be larger) aligns the nematogens more effectively. Whilst the *lengthscale* of aligned regions $\xi_{G,d}$ may increase, the *magnitude* of $\mathcal{C}^G$, which governs the intensity of the alignment locally, decreases [17]. By contrast, for $\mathrm{H}_\mathbf{0} > \mathrm{H}^{(c)}$ (i.e., the *strong disorder regime*), the (weak disorder) simple decay of the correlators can give way to oscillatory decay, as we now discuss. Regardless of T, $\mathcal{C}^T$ oscillates, whereas $\mathcal{C}^G$ only does for sufficiently small T. The oscillation wavelengths $\xi_{T/G,o}$ are determined via the radii of the shells in wavevector space on which the corresponding correlators are maximal. Thus, we arrive at an explicit (and, notably, T-independent) formula $\xi_{T,o} = \xi_L/\sqrt{2\ln(\mathrm{H}_\mathbf{0}/\mathrm{H}^{(c)})}$ and an implicit one for $\xi_{G,o}$, viz.,

$$1 + (\xi_N/\xi_{G,o})^2 + 4(\xi_N/\xi_L)^2 - (\mathrm{H}_\mathbf{0}/\mathcal{A}t)e^{-\xi_L^2/2\xi^2_{G,o}} = 0.$$

The value of $\xi_{T/G,d}$ in this strong-disorder regime, given in Table I, is estimated via the widths of the peaks of $\mathcal{C}^{T/G}$. Upon decreasing ξ_L at fixed ξ_N, the value of $\xi_{T,d}$ tends to ξ_L from above, indicating that the network is limiting the range over which the thermal nematic fluctuations are correlated; on the other hand, $\xi_{G,d}$ remains at the scale of ξ_L, indicating that the range of coherent nematic alignment is circumscribed by the network's typical localization length.

Oscillatory behavior can be regarded as the resolution of the interplay of two energetic costs of fluctuations. The cost of creating local nematic order *via rotations* of the nematogens is smaller for long-wavelength fluctuations. By contrast, the cost of creating nematic order *via local segregation* of nematogens according to their preferred

orientation is smaller for short-wavelength fluctuations (which is a reflection of the short-lengthscale liquidity of the network). When the former mode dominates for all wavelengths, long-wavelength fluctuations are the most probable and, hence, correlations decay without oscillation. When the disorder is strong enough, however, the latter mode drives the most probable fluctuations to a finite wave-vector and, hence, correlations oscillate as they decay. (Such behavior is analogous to the micro-phase separation in cross-linked polymer blends. [18, 19])

Having considered the behaviors of $\mathcal{C}^T$ and $\mathcal{C}^G$ for systems prepared at high temperatures, we now consider the corresponding behavior for systems prepared at $t_p < T\mathrm{H}_{\mathbf{0}}/T_p\mathcal{A}^0$, so that the local nematic order present immediately post cross-linking is spatially correlated over distances larger than ξ_L [20]. As one can see from Eq. (9a), the behavior of $\mathcal{C}^T$ is unchanged, undergoing simple decay in real space at weak disorder but oscillatory decay at strong disorder. Conversely, $\mathcal{C}^G$ exhibits behavior qualitatively different from that of a system prepared at $t_p \gg T\mathrm{H}_{\mathbf{0}}/T_p\mathcal{A}^0$, because it now receives its dominant contribution from the memorization of $\mathbf{Q}^0$. Specializing to $t \approx t_p$ and for wavelengths larger than ξ_L, we see from Eq. (9b) that $\mathcal{C}^G$ is approximately given by

$$\mathcal{C}^G_{\mathbf{k}} \approx \mu_D \left(\frac{T}{T_p}\right)^2 \frac{T_p}{\mathcal{A}^0 t_p + \mathcal{L}^0 k^2}, \qquad (10)$$

i.e., it is proportional to the correlator of the thermal nematic fluctuations immediately post cross-linking. This indicates that the pattern of these thermal fluctuations has been *faithfully memorized* by the network.

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[10] See, e.g., P.-G. de Gennes and J. Prost, *The Physics of Liquid Crystals*, 2nd edn (Clarendon Press, Oxford, 1993). As our primary aim is uncover the qualitative physics encoded in Eq. (3), we have neglected another possible gradient term, viz. $\mathcal{L}' \int_{\mathbf{k}} \sum_{d,d',d''=1}^{D} k_{d'} Q_{dd'}(\mathbf{k})\, k_{d''} Q_{dd''}(-\mathbf{k})$, where $\mathcal{L}'$ is the generalized stiffness corresponding to this term. Including this gradient term in Eq. (3) will result, for example, in the five components of the nematic tensor $\mathbf{Q}$ not all having the same value of the oscillation wavelength $\xi_{G/T,o}$ at strong disorder. However, the inclusion would not change our results *qualitatively*, including our central result, viz., that spatially oscillatory decay arises for the glassy and thermal nematic correlators, provided the disorder is sufficiently strong.
[11] The reduced measurement temperature, t, is defined to be $(T - T^*)/T^*$, where T^* is the spinodal temperature for the spatially homogeneous isotropic-to-nematic transition in the nematic liquid. The reduced preparation temperature is defined via $t_p \equiv (T_p - T^*)/T^*$.
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[13] In the high-temperature regime (which is our concern in this paper), in which local nematic order is induced by the random field $\mathbf{M}$, incorporating the effects of local elastic deformations would downward-renormalize t as well as $\mathcal{L}$, but will not change the qualitative picture presented in the paper. This is demonstrated in [9].
[14] Although we are focusing on the scalar aspects of the correlators, it is straightforward to reconstruct the full structure of the corresponding fourth-rank tensors, by appending suitable isotropic tensor factors constructed from Kronecker deltas.
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[17] This can be seen by computing the value of $\mathcal{C}^G(\boldsymbol{r} = \mathbf{0})$, which is approximately given by $T\mu_D \mathrm{H}_{\mathbf{0}} \xi_N / 4\pi\mathcal{L}^2$ at weak disorder, and noting that $\mathrm{H}_{\mathbf{0}}$ varies as ξ_L^{-4}.
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[20] This regime can be reached in IGNEs by imposing a sufficiently large density of crosslinks, such that the typical localization length of the network becomes comparable to or smaller than the nematic correlation length of the isotropic liquid of nematogens just prior to cross-linking. We note that in addition to IGNEs, the result that we have obtained for this regime also describes the strong memory effect present in nematic elastomers that

have been prepared in the nematic state, at least qualitatively [3].