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Issues of replica-symmetry breaking for the amorphous solid state of vulcanized macromolecules

Paul M Goldbart†§ and Annette Zippelius‡||

† Department of Physics, Materials Research Laboratory and Beckman Institute, University of Illinois at Urbana-Champaign, Urbana, IL 61801, USA

‡ Institut für Theoretische Physik, Georg August Universität, D 37073 Göttingen, Germany

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Abstract. The statistical mechanics of vulcanized (i.e. permanently randomly crosslinked) macromolecular matter can be formulated, using the replica technique, as the $n \rightarrow 0$ limit of a theory containing $n + 1$ coupled replicas. Within the framework of a replica-symmetric variational mean-field approach, this theory describes an equilibrium phase transition, upon sufficient vulcanization, from a liquid to an amorphous solid state. We consider a natural extension of this framework, which admits the possibility of the spontaneous breaking of replica symmetry, and find that—at least at the mean-field level—replica symmetry appears to remain intact. We discuss the physical origin of this absence of replica-symmetry breaking, as well as a possible strategy for a theoretical refinement that may yield replica-symmetry breaking.

In a recent letter [1] (see also [2–4]), a statistical mechanical theory has been presented of the equilibrium phase transition, from the liquid state to the amorphous solid state, that macromolecular networks exhibit upon the introduction of a sufficient density of permanent random crosslinks (i.e. upon sufficient vulcanization). The central feature of the theory presented in [1] is a one-parameter variational hypothesis for the order parameter appropriate to the transition at hand, namely, a type of Edwards–Anderson order parameter [5, 6] capable of detecting the random static density fluctuations that emerge at this solidification transition [4, 7]. The variational parameter may be regarded as a localization length for the monomers: it is infinite in the liquid state and becomes finite, continuously, at the transition. (A closely related description of covalently bonded random atomic networks—i.e. structural glasses—has also recently been developed [8].) The purpose of the present paper is to extend the investigation begun in [1] by investigating the possibility of the spontaneous breaking of replica symmetry (RS) in the equilibrium amorphous solid state of randomly crosslinked macromolecular networks and to explore the implications of replica-symmetry breaking (RSB) for the structure of the space of equilibrium states.

This paper is organized as follows. We begin by briefly sketching the replica mean-field theory approach to randomly crosslinked macromolecular networks and outline an RS variational hypothesis for the order parameter. (The reader is referred to [1], and to references therein, for further details.) We then indicate how the variational hypothesis may be broadened so as to allow for the spontaneous breaking of RS and, following this, we present a specific pattern for breaking the RS. One of the novel aspects of these issues

§ e-mail address: goldbart@uiuc.edu

|| e-mail address: annette@theo-phys.gwdg.de

becomes apparent from the recognition that, in the replica approach, randomly crosslinked macromolecular networks are described by the $n \rightarrow 0$ limit of a theory involving $n + 1$ replicas, rather than the usual n replicas [2]. Next, we examine the consequences of interpreting this pattern of RSB in terms of pure equilibrium states and, finally, we examine the consequences of implementing this pattern at the one-step level in the specific context of a variational mean-field theory of randomly crosslinked macromolecular networks. In fact, we shall find that the system does *not* take advantage of RSB, at least in the form presented here. We conclude with a discussion on the physical origin of the absence of RSB shown by the present mean-field theory and with a conjecture regarding a more elaborate description of randomly crosslinked macromolecular networks that we anticipate might exhibit RSB.

The system to be considered consists of N macromolecules of arclength L and persistence length ℓ moving in a d -dimensional volume V . The macromolecules are labelled by the index $i = 1, \dots, N$ and the configuration of the system is described by the collection of spatial trajectories $\{\sqrt{L\ell/d}c(s)\}_{i=1}^N$, where $0 \leq s \leq 1$. (In other words, we measure spatial distances in units proportional to the RMS end-to-end distance of a free macromolecule; arclengths in units of the total arclength and energies in units such that $k_B T = 1$.) We model the uncrosslinked system using the Edwards Hamiltonian [9, 10] and, following Deam and Edwards [2], adopt a crosslink distribution that reflects the correlations present in the uncrosslinked macromolecular liquid, particularly the integrity of the macromolecules and the space-filling nature of the typical configurations of the liquid. The permanent random crosslinks play the role of quenched random variables; their mean density is controlled by the parameter μ^2 .

By applying the replica technique and introducing the order-parameter field $\Omega_{\hat{k}}$, we can formulate the problem in terms of an effective Hamiltonian $f_n\{\Omega\}$ for a single (replicated) macromolecule

$$nf_n\{\Omega\} = \frac{\mu^2}{2Vn} \sum_{\hat{k}} |\Omega_{\hat{k}}|^2 - \ln \left\langle \exp \left(\frac{\mu^2}{2Vn} \sum_{\hat{k}} \Omega_{\hat{k}} \int_0^1 ds e^{-i\hat{k} \cdot \hat{c}(s)} + \text{cc} \right) \right\rangle_{n+1}^W. \quad (1)$$

Collections of $(n + 1)$ -fold replicated d -dimensional vectors are denoted by hatted variables; e.g. \hat{c} denotes $\{c^0, \dots, c^n\}$. Angle brackets $\langle \dots \rangle_{n+1}^W$ denote averaging with respect to the $(n + 1)$ -fold replicated Wiener measure for a single (replicated) macromolecule.

The expectation value $\langle \Omega_{\hat{k}} \rangle$, taken with the Boltzmann weight $\exp(-Nnf_n\{\Omega\})$, is related to the disorder average of the static density fluctuations

$$\langle \Omega_{\hat{k}} \rangle = \frac{1}{N} \sum_{i=1}^N \int_0^1 ds \langle \exp(i\hat{k} \cdot \hat{c}_i(s)) \rangle. \quad (2)$$

When at least two of the $(n + 1)$ -component vectors in \hat{k} are non-zero, then $\langle \Omega_{\hat{k}} \rangle$ is the order parameter that distinguishes between liquid and amorphous solid states, detecting the emergence of random static density fluctuations [4]. This order parameter characterizes *all* moments of the distribution of monomer-specific densities, not solely the second moment. A mean-field description follows by considering the effective Hamiltonian (1) in the stationary-point approximation. Then, the disorder-averaged free energy per macromolecule f (relative to the free energy of the uncrosslinked liquid) is given by $f = \lim_{n \rightarrow 0} f_n$.

In [1], we discussed in detail the RS variational hypothesis

$$\Omega_{\hat{k}} = \delta_{\sum_{\alpha=1}^n k^\alpha, 0} \exp(-\xi^2 \hat{k}^2 / 2) \quad (3)$$

where $\hat{k}^2 \equiv \sum_{\alpha=0}^n |k^\alpha|^2$. The variational parameter ξ can be interpreted as a localization length: it is infinite in the liquid state and finite in the amorphous solid state. The Kronecker-delta factor encodes the physical notion that the amorphous solid state is macroscopically translationally invariant, despite the microscopic translational symmetry breaking that occurs through the emergence of random static density fluctuations. *Inter alia*, it is shown in [1] that, within the context of this hypothesis, a phase transition occurs between the liquid and amorphous solid states at a critical value of μ^2 , namely, $\mu^2 = \mu_c^2 \approx 1.59$. (In recent work we have introduced a broader RS variational hypothesis in which the ordered state is characterized by a *distribution* of localization lengths. This more elaborate hypothesis turns out to yield a stationary point of the effective Hamiltonian [11].)

To allow for the possibility of RSB, it is natural to generalize the variational hypothesis (3) by adopting the form

$$\Omega_{\hat{k}} = \delta \sum_{\alpha=0}^n k^\alpha \cdot 0 \exp \left(-\frac{1}{2} \tilde{\Gamma} \sum_{\alpha=0}^n |k^\alpha|^2 + \frac{1}{2} \tilde{\Gamma} \sum_{\alpha,\beta=0}^n \hat{\Gamma}^{\alpha\beta} k^\alpha \cdot k^\beta \right) \quad (4)$$

with $\tilde{\Gamma}$ non-negative and with the matrix $\hat{\Gamma}$ satisfying the constraints that its diagonal elements all vanish and that its off-diagonal elements average to zero, i.e. $\sum_{\alpha \neq \beta} \hat{\Gamma}^{\alpha\beta} = 0$. To express the hypothesis in this particular form turns out to be convenient. One should regard $\tilde{\Gamma}$ as encoding RS information and $\hat{\Gamma}^{\alpha\beta}$ as encoding information concerning RSB. Related hypotheses have been invoked in the context of systems with n replicas [12].

Inserting the variational hypothesis (4) into the effective Hamiltonian (1), expanding perturbatively in $\tilde{\Gamma}^{-1}$ and retaining terms to all orders in $\hat{\Gamma}$, we obtain the mean-field free energy (valid for small n)

$$(2/d) f_n = \tau(\mu^2) \{ \ln(2\pi e \tilde{\Gamma} / V^{2/d}) + E_n \} - (\mu^2/2) F_n + h(\mu^2) \{ 1 + G_n \} \frac{1}{6\tilde{\Gamma}} + \mathcal{O}(\tilde{\Gamma}^{-2}) \quad (5)$$

where

$$1 + nE_n \equiv \det(\hat{1} - \hat{\Gamma}) \quad (6a)$$

$$(1 + n) + nF_n \equiv \sum_{\alpha,\beta=0}^n (\hat{1} - \hat{\Gamma})^{-1} |^{\alpha\beta} \quad (6b)$$

$$(1 + n) + nG_n \equiv \text{tr}(\hat{1} - \hat{\Gamma})^{-1} \quad (6c)$$

and where $h(x) \equiv \{e^{-x} - (1 - x)\}$, $\tau(x) \equiv h(x) - (x/2)$ and $\hat{1}$ is the $(n + 1) \times (n + 1)$ identity matrix. It should be noted that for any RSB pattern in which all replicas remain equivalent, such as the one to be discussed below, it can readily be established that $F_n = 0$.

We now make a specific assumption about the form of the RSB pattern. Consider the $(1 + n) \times (1 + n)$ matrix $\hat{\Gamma}$. Partition the $(1 + n)$ replicas into $(1 + n)/(1 + \epsilon n)$ equivalent groups, each containing $(1 + \epsilon n)$ replicas. The parameter ϵ should be regarded as interpolating between (1×1) and $(1 + n) \times (1 + n)$ matrices; it characterizes the pattern of RSB and satisfies the inequalities $0 \leq \epsilon \leq 1$. Next, to the off-diagonal elements of the matrix $\hat{\Gamma}$ associated with all pairs (α, β) in the *same* partition (i.e. for elements of $\hat{\Gamma}$ in the *diagonal* blocks), assign the value $-(1 - \epsilon)g$; to the elements of $\hat{\Gamma}$ associated with pairs (α, β) in *distinct* partitions (i.e. for elements of $\hat{\Gamma}$ in the *off-diagonal* blocks), assign the value ϵg . (The matrix elements are not independent because of the constraint that their

average over the matrix $\hat{\Gamma}$ is zero.) The parameter g should be regarded as characterizing the strength of RSB. As the block size depends explicitly on n , this one-step RSB pattern is a slight variant of the Parisi scheme [13, 5, 6], the variance arising from the need to explore the neighbourhood of (1×1) matrices rather than (0×0) . This particular scaling of the block size $(1 + \epsilon n)$ with the number of replicas $(1 + n)$ is chosen for three reasons: first, it leads to a natural interpretation of RSB in terms of the distribution of overlaps between pure equilibrium states; second, it leads to the free energy having a sensible $n \rightarrow 0$ limit; and third, it can be iterated an arbitrary number of times.

The iterability of the RSB pattern can usefully, if heuristically, be illustrated using the language of group theory [14]. Let \mathcal{S}_n denote the group of permutations of n objects. Then the present RSB pattern can be depicted as follows

$$\mathcal{S}_{1+n} \rightarrow (\mathcal{S}_{1+\epsilon n})^{\otimes \frac{1+n}{1+\epsilon n}} \hat{\otimes} \mathcal{S}_{\frac{1+n}{1+\epsilon n}} \quad (7)$$

where the symbol on the left-hand side indicates the (permutation aspects of the) order-parameter symmetry group in the absence of RSB and the symbol on the right-hand side indicates the residual symmetry under the present RSB scheme [15]. Taking the $n \rightarrow 0$ limit gives the symmetry-breaking pattern

$$\mathcal{S}_1 \rightarrow \mathcal{S}_1 \hat{\otimes} \mathcal{S}_1 \quad (8)$$

which shows that the broken symmetry group contains, as a subgroup, a copy of the unbroken symmetry group, so that the pattern of symmetry breaking can be iterated indefinitely. It should be noted that invoking the Parisi pattern of RSB in its usual form (i.e. choosing the block size not to have any explicit dependence on n) does not lead to a readily iterable pattern of RSB in the present context. Iterating the present scheme K times corresponds to choosing a sequence of variational parameters

$$1 \equiv \epsilon_0 \geq \epsilon_1 \geq \dots \geq \epsilon_k \geq \dots \epsilon_K \geq \epsilon_{K+1} \equiv 0 \quad (9a)$$

governing the sequence of block sizes $m_k = 1 + \epsilon_k n$ and a sequence of values

$$\Gamma_0 \geq \Gamma_1 \geq \dots \geq \Gamma_k \geq \dots \Gamma_{K-1} \geq \Gamma_K \quad (9b)$$

for the corresponding elements of the matrix $\hat{\Gamma}$, satisfying the constraint

$$\sum_{k=0}^K (\epsilon_k - \epsilon_{k+1}) \Gamma_k = 0.$$

Let us now turn to the interpretation of RSB. By using the clustering properties of expectation values in pure equilibrium states, it can readily be shown [16, 5, 4] that

$$P_{\{kk'\}}(q) \equiv \left[\sum_{\sigma\sigma'} w^\sigma w^{\sigma'} \delta \left(q - Q_{kk'}^{\sigma\sigma'} \right) \right] \simeq \lim_{n \rightarrow 0} \frac{2}{(n+1)n} \sum_{0 \leq \alpha < \beta \leq n} \delta \left(q - w_{\{kk'\}}^{\alpha\beta} \right). \quad (10)$$

Here, σ labels the pure equilibrium states, w^σ denotes the normalized Boltzmann weight of the state σ , $\langle \dots \rangle^\sigma$ denotes expectation values taken within the state σ ,

$$Q_{kk'}^{\sigma\sigma'} \equiv \frac{1}{N} \sum_{i=1}^N \int_0^1 ds \langle e^{zik \cdot c_i(s)} \rangle^\sigma \langle e^{ik' \cdot c_i(s)} \rangle^{\sigma'} \quad (11)$$

is the (unsymmetrized [4]) overlap (at wavevectors \mathbf{k} and \mathbf{k}') of the pair of states σ and σ' and the square brackets $[\dots]$ denote averaging over the distribution of crosslink number and positions. Thus, $P_{\{\mathbf{k}\mathbf{k}'\}}(q)$ has the meaning of the disorder-averaged Boltzmann-weighted distribution of (symmetrized) overlaps (at wavevectors \mathbf{k} and \mathbf{k}') between pairs of pure equilibrium states. How to compute this distribution using the replica technique is specified on the right-hand side of equation (10), in which an average is taken over all equivalent stationary points generated by the unbroken symmetry group of the theory. The value of the order parameter $\Omega_{\hat{k}}$ features via the quantity $\omega_{\mathbf{k}\mathbf{k}'}^{\alpha\beta}$, which is defined to be the value of $\Omega_{\hat{k}}$ when all wavevectors in \hat{k} vanish except the α th (which equals \mathbf{k}) and the β th (which equals \mathbf{k}').

Distributions of overlaps of more than two pure states can also be considered, leading to results similar to those given above for pairs of states. The information necessary to compute such distributions is contained in the values of $\Omega_{\hat{k}}$ when more than two wavevectors in \hat{k} are non-zero. In addition, one can consider distributions of overlaps between pure states arising from the decomposition of the physical Gibbs ensemble and the analogues of pure states arising from the decomposition of the ensemble used to generate the crosslink distribution. In situations such as the present one, in which the crosslink-distribution-generating ensemble is identical to the physical ensemble, the unbroken symmetry group of the replica theory is \mathcal{S}_{n+1} . Then, there is no distinction between those overlap distributions involving only physical pure states and those also involving their analogues arising from the decomposition of the crosslink-distribution-generating ensemble. It would, however, also be of interest to consider systems in which the physical and crosslink-distribution-generating ensembles are distinct, e.g. because crosslinking is undertaken at a different temperature or in the presence of a solvent of different quality. Then, the unbroken symmetry group of the replica theory would instead be \mathcal{S}_n and one should anticipate a distinction between overlap distributions involving only physical pure states and those also involving the analogues of pure states arising from the decomposition of the crosslink-distribution-generating ensemble.

Within the context of the variational hypothesis (4), the present pattern of RSB (implemented at the one-step level) leads to the overlap distribution

$$P_{\{\mathbf{k}\mathbf{k}'\}}(q) \simeq (1 - \epsilon)\delta\left(q - \delta_{\{\mathbf{k}\mathbf{k}'\},0} e^{-k^2\bar{\Gamma}(1+\epsilon g)}\right) + \epsilon\delta\left(q - \delta_{\{\mathbf{k}\mathbf{k}'\},0} e^{-k^2\bar{\Gamma}(1-(1-\epsilon)g)}\right) \quad (12)$$

which evidently is sensible (and yet potentially non-trivial) in the $n \rightarrow 0$ limit and which is acceptable as a probability distribution, provided that $0 \leq \epsilon \leq 1$. It should be emphasized that the good features mentioned here arise as a consequence of the explicit n -dependence conferred upon the sizes of the blocks in the present scheme. If the RSB pattern were iterated beyond the one-step level then the distribution would contain more than two delta-functions, their weights remaining non-negative but potentially non-trivial by virtue of the adopted scaling of the (sequence of) block sizes with n

$$P_{\{\mathbf{k}\mathbf{k}'\}}(q) \simeq \sum_{j=0}^K (\epsilon_j - \epsilon_{j+1})\delta\left(q - \delta_{\{\mathbf{k}\mathbf{k}'\},0} e^{-k^2\bar{\Gamma}(1+\Gamma_j)}\right). \quad (13)$$

RSB can be pictured qualitatively as inducing some dispersion of the overlaps: the overlaps of distinct states can be interpreted as yielding larger (mutual) localization lengths than do the self-overlaps of states.

By applying this one-step RSB scheme to the effective Hamiltonian (5), expanding perturbatively to second order in $\bar{\Gamma}^{-1}$ and taking the $n \rightarrow 0$ limit (and omitting irrelevant

constants), we find that the mean-field free energy f is given by

$$(2/d)f = \tau(\mu^2)\{\ln Q^{-1} + (1 - \epsilon)\ln(1 + J)\} + h(\mu^2)\{1 - (1 - \epsilon)J(J + 1)^{-1}\}Q \\ + k(\mu^2)\{1 - (1 - \epsilon)J(J + 2)(J + 1)^{-2}\}Q^2 + \mathcal{O}(Q^3) \quad (14)$$

where

$$Q^{-1} \equiv 6\tilde{\Gamma}\{1 - (1 - \epsilon)g\} \quad (15a)$$

$$J \equiv g\{1 - (1 - \epsilon)g\}^{-1} \quad (15b)$$

$$k(x) \equiv e^{-x} \sum_{r=2}^{\infty} \frac{x^r}{r!} \frac{r-1}{5r} (2r^2 + 2r + 3) \quad (15c)$$

and where we have exchanged the set of order parameters $\{\tilde{\Gamma}, g, \epsilon\}$ for the set $\{Q, J, \epsilon\}$. For $\tau \leq 0$, (i.e. $\mu^2 \leq \mu_c^2 \approx 1.59$) f has only the trivial stationary point $\tilde{\Gamma} = \infty$, corresponding to the liquid (i.e. delocalized) state. At $\tau = 0$ (i.e. $\mu^2 = \mu_c^2$) there is a continuous transition from the liquid state to an amorphous solid state, in which the monomers are localized, having $\tilde{\Gamma} < \infty$. For $\tau > 0$ (i.e. $\mu^2 > \mu_c^2$) an RS stationary point of f corresponding to the amorphous solid state is readily found, at least in the vicinity of the transition (i.e. for $\tau \ll 1$), e.g. by setting J to zero and requiring that f be stationary with respect to Q . This solution corresponds to the state described in [1].

To search for RSB stationary points of f in the vicinity of the transition, we first assume the power-series expansions in τ

$$hQ = Q_1\tau + Q_2\tau^2 + \dots \quad (16a)$$

$$J = J_1\tau + \dots \quad (16b)$$

with ϵ arbitrary. The conditions that f be stationary with respect to Q and to J then yield only one stationary point, an RS stationary point, at which $g = 0$ (or, equivalently, $\epsilon = 0$ or 1) and $\tilde{\Gamma}^{-1} \approx 6\tau(1 + h^{-2}k\tau)$; we do not find an RSB stationary point. Next, we consider the possibility of a stationary point that corresponds to the situation in which the overlaps between all pairs of distinct states vanish but the self-overlaps share a common non-zero value. This case also yields an RS solution only, the weight of the putative (zero-valued) non-self-overlaps being zero. Finally, we consider the possibility of RSB (also at the one-step level) according to the scheme adopted by Seung *et al* [17] in the specific context of supervised learning by neural networks: with this scheme, once again, we do not find an RSB stationary point†.

Genuine physical networks of randomly crosslinked macromolecules seem, *a priori*, to be likely candidate systems for exhibiting RSB. However, the present calculation appears to favour an RS state. We conjecture that the origin of the absence of RSB lies in the neglect of the (crosslink-renormalized) excluded-volume interaction, which results from the mean-field treatment. Thus, the thermodynamic barriers necessary to delineate between distinct pure equilibrium states have been omitted from the theory and RSB is not realized at the mean-field level. It would, therefore, be most interesting to enquire whether or not

† For systems having the full S_{n+1} permutation symmetry, the pattern of RSB considered in [17] leads to the unattractive feature that the probabilistic interpretation of RSB, e.g. as in equation (12), suffers from the occurrence of negative weights.

the incorporation of interactions—and thus barriers—does indeed lead to the spontaneous breaking of RS. One strategy for accomplishing this, which is a particularly natural extension of the present framework, is to treat the excluded-volume interaction at the Gaussian level, following the collective coordinate method of Edwards [18]. This entails a somewhat more elaborate calculation than that presented here, a calculation on which we intend to report in the near future [19].

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