

Translational invariance in models for low-temperature properties of glasses

Reimer Kühn and Jens Urmann

Institut für Theoretische Physik, Universität Heidelberg, Philosophenweg 19, 69120 Heidelberg, Germany

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Abstract. We report on a refined version of our spin-glass-type approach to the low-temperature physics of structural glasses. Its key idea is based on a Born–von Karman expansion of the interaction potential about a set of reference positions in which glassy aspects are modelled by taking the harmonic contribution within this expansion to be random. Within the present refined version the expansion at the harmonic level is reorganized so as to respect the principle of global translational invariance. By implementing this principle, we have for the first time a mechanism that fixes the distribution of the parameters characterizing the local potential energy configurations responsible for glassy low-temperature anomalies solely in terms of assumptions about interactions at a microscopic level.

1. Introduction

The present contribution is intended to further explore and refine our spin-glass way of looking at low-temperature anomalies in glasses developed earlier [1, 2]. The term ‘low-temperature anomalies’ (LTA) refers to a set of observations according to which a number of thermodynamic and transport properties of glassy and amorphous systems have been found to differ drastically and unexpectedly from those of their crystalline counterparts [3], when the temperature is lowered to a few K.

In particular, below approximately 1 K the specific heat of glassy materials has been found [3] to scale approximately linearly with temperature, $C \sim T$, while the corresponding scaling for the thermal conductivity κ is approximately quadratic, $\kappa \sim T^2$. Both findings contrast with the T^3 -behaviour of these quantities in crystals. Between 1 K and approximately 20 K the thermal conductivity displays a plateau and continues to rise as the temperature is further increased. The specific heat also changes its behaviour in the 1–20 K regime. It exhibits a peak if displayed in C/T^3 plots, signifying an excess density of states in that energy range, which is often referred to as the Bose peak. LTA are remarkable both for their ubiquity, and for their peculiar pattern of universality and the absence thereof in various temperature ranges [3, 4]; for reviews, see [5, 6] and the recent collection [7].

The anomalous properties of glasses at low temperatures are usually attributed to the existence of a broad range of localized low-energy excitations in amorphous systems—excitations not available in crystalline materials. At energies below 1 K, these are thought to be tunnelling excitations of single particles or small groups of particles in double-well configurations of the potential energy (DWPs). This is the main ingredient of the phenomenological so-called standard tunnelling model (STM), independently proposed by

Phillips [8] and by Anderson *et al* [9]. As a second ingredient of the STM, it is supposed that the local DWPs in amorphous systems are random, and specific assumptions concerning the distribution of the parameters characterizing them are advanced to describe the experimental data below 1 K [8, 9]. Excitations at energies between 1 K and 20 K responsible for the boson peak and (via resonant scattering of phonons) presumably for the plateau in the thermal conductivity are believed to be of a different nature, namely localized vibrations in anharmonic single-well configurations of the potential energy (SWPs). The existence of such single-well potentials is the main additional assumption of the likewise phenomenological soft-potential model [10, 11] (SPM). Within the SPM it is supposed that locally the potential energy surface (along some reaction coordinate) can be described by certain fourth-order polynomials, with coefficients distributed in a specific way so as to comprise both DWPs and SWPs, the former giving rise to tunnelling systems, the latter to localized vibrations.

In both the STM and the SPM, a weak coupling between localized excitations and extended (phonon) modes is assumed to describe the phenomenology of heat transport and the anomalous acoustic or dielectric properties of glasses at low temperatures [12].

Although the STM and the SPM describe the phenomenology of glassy LTA reasonably well, the situation cannot be considered entirely satisfactory.

To begin with, neither model accounts for a *mechanism* that would explain *how* the required local potential energy configurations would arise, and how they would do so with the required statistics. Indeed, as we have discussed elsewhere [13], the assumptions concerning the distributions of parameters characterizing the local potential energy configurations in either phenomenological approach are at best only partially plausible. Moreover, neither model can explain the considerable degree of universality of the LTA, or the observed absence of universality at intermediate temperatures. As phenomenological models for LTA, they also have little to say about relations between low-temperature phenomena and the physics at the glass transition. Finally, there is growing experimental evidence that things may go wrong with the assumptions of the STM (and the SPM) as the temperature is lowered into the mK regime. Let us mention:

- (i) the unexpectedly rapid decay of coherent echoes in glasses [14],
- (ii) the temperature dependence of acoustic attenuation and dispersion [15],
- (iii) unexpected features in the long-time behaviour of hole-burning experiments [16],
- (iv) the unreasonably large value of 3 mK for the minimal tunnelling matrix element deduced from specific heat data [17] and dielectric measurements [18], and finally
- (v) the recently reported evidence for a macroscopic quantum state of tunnelling systems in glasses below 5 mK [19].

These all point to the necessity for a better understanding of interaction effects in glasses at (very) low temperatures.

It is with these observations in mind that our model-based spin-glass way of looking at low-temperature anomalies in glasses attempts to fill a gap, and we believe it to carry considerable potential for clarifying at least some of the unresolved issues that have emerged lately.

Our approach is based on a Born–von Karman expansion of the interaction potential of a glassy system about a set of reference positions, in which glassy aspects are modelled by taking the harmonic contribution within this expansion to be random. We derive the justification for such a procedure from the observation of universality: since the low-temperature anomalies observed in amorphous systems are apparently to a large extent insensitive to the detailed form of the interaction, *any* interaction might be taken as a starting point, as long as it does give rise to a glassy low-temperature phase.

The approach leads to a class of models of spin-glass type which exhibit both glassy low-temperature phases, and double- and single-well configurations in their potential energy. The distribution of parameters characterizing the local potential energy configurations can be *computed*, and differ from those assumed in the standard tunnelling model and its variants. Still, the low-temperature anomalies characteristic of amorphous systems are reproduced, and we are able to distinguish properties which can be expected to be universal from those which cannot.

We have organized our material as follows. In section 2 we briefly describe the original variant of our approach and describe the main results derived from it. Section 3 introduces a refined version in which the expansion at the harmonic level is modified to respect the principle of global translational invariance, from which we derive new insights concerning the relation between the original particle–particle interaction and the distribution of parameters characterizing the local potential energy configurations. Section 4 is devoted to main results, and section 5 to concluding remarks.

2. The spin-glass approach—original set-up

We begin by briefly describing the main ingredients of our original model, referring the reader to [1, 2, 13, 20] for details, numerical results, and illustrations.

In the original variant of our spin-glass approach to glassy low-temperature anomalies, we suggested considering the following Hamiltonian as a candidate for the description of a set of particles forming a glassy system:

$$\mathcal{H} = \sum_{i=1}^N \frac{p_i^2}{2m} + U_{\text{int}}(\{u_i\}) \quad (1)$$

with an interaction energy given by

$$U_{\text{int}}(\{u_i\}) = -\frac{1}{2} \sum_{i \neq j} J_{ij} u_i u_j + \sum_i G(u_i) \quad (2)$$

in which glassy aspects are modelled by taking the J_{ij} to be random. On-site potentials of the form

$$G(u) = \frac{a_2}{2} u^2 + \frac{a_4}{4!} u^4 \quad (3)$$

are included to stabilize the system as a whole. Through the harmonic term in G , the parameter a_2 controls the number of modes that are unstable at the harmonic level of description. The parameter a_2 may be fixed, or chosen according to some non-degenerate distribution as well (see below).

The description is in terms of *localized* degrees of freedom, i.e., the u_i are interpreted as deviations of particle positions from a given set of reference positions as in a Born–von Karman expansion known from the dynamical theory of crystalline solids. Thus, the system is assumed to be already in a solid state, and no attempt to provide a faithful description of the liquid phase is made.

The random interactions are chosen in such a way that the system can be analysed within replica mean-field theory, e.g., as in the SK model [21]. This implies that the potential energy surface of the system can be represented as a sum of effective independent single-site potential energies $U_{\text{eff}}(u_i)$ containing random parameters:

$$U_{\text{int}}(\{u_i\}) \longrightarrow \sum_i U_{\text{eff}}(u_i). \quad (4)$$

In the replica-symmetric (RS) approximations these potentials are of the form [1, 2]

$$U_{\text{eff}}(u) = -h_{\text{eff}}u - \frac{1}{2}J^2Cu^2 + G(u) \quad (5)$$

with

$$h_{\text{eff}} = h_{\text{RS}} = J_0p + J\sqrt{q}z \quad (6)$$

and $C = \beta(q_d - q)$. Here p denotes a macroscopic polarization, and q_d and q the diagonal and off-diagonal entries of the RS Edwards–Anderson order parameter matrix. Apart from randomness that may be present in $G(u)$, the effective single-site potentials contain a single random parameter, namely the Gaussian-distributed effective fields h_{eff} having mean J_0p and variance J^2q . The parameters p , q , and C characterizing the $U_{\text{eff}}(u)$ ensemble are determined self-consistently through a set of saddle-point equations[†].

The model exhibits non-ergodic low-temperature phases, which may be glassy or polarized, depending on the parameters J_0 and J ; the glass transition temperature (or the temperature of the transition into the polarized phase) depends on a_2 as well. Replica-symmetry breaking (RSB) occurs at low temperatures and small a_2 , and has been analysed within a one-step replica-symmetry-breaking (1RSB) approximation in [2]. Its main effect is to modify the h_{eff} -distribution, and in particular to reveal correlations between the h_{eff} and whatever randomness one might have considered for the $G(u)$.

The relevance of these results for glassy LTA derives from the fact that the $U_{\text{eff}}(u)$ acquire a harmonic term $-\frac{1}{2}J^2Cu^2$ —entirely of collective origin—that renormalizes the local harmonic restoring force produced by $G(u)$. Hence for

$$J^2C > a_2 \quad (7)$$

the total harmonic contribution to $U_{\text{eff}}(u)$ becomes convex downward near the origin $u = 0$, so for sufficiently small h_{eff} the effective single-site potential $U_{\text{eff}}(u)$ attains a DWP form, which is of collective origin.

The existence within the ensemble of effective single-site potentials of a spectrum of DWPs with a broad distribution of asymmetries is mainly responsible for the appearance of glassy LTA via low-energy tunnelling excitations with a virtually constant density of states (DOS) at low energies, giving rise to the well-known linear temperature scaling of the specific heat at low temperatures. Higher-order excitations in DWPs and quasi-harmonic excitations in SWPs give rise to a peak in the DOS, and consequently to a Bose peak. These results were presented and discussed in detail in [1, 2, 13].

Let us close this section with a few remarks concerning the distribution of parameters in the effective single-site potentials.

If we choose the parameter a_2 in (2), (3) to be fixed and non-random, the effective single-site potentials $U_{\text{eff}}(u_i)$ contain only a single random parameter, namely the effective field h_{eff} , in contrast to the STM and the SPM, both of which assume two randomly varying parameters. Indeed, whereas the low-temperature specific heat comes out correctly without a randomly varying harmonic contribution to $G(u)$, dynamic properties such as ultrasound attenuation do require a broad a_2 -distribution in order to be reproduced correctly [20, 22]. While an assumption of this kind is natural within our approach in view of the fact that the harmonic contribution to $G(u)$ might have been omitted in favour of diagonal entries J_{ii} in the (random) interaction matrix, it still creates the (awkward) need for an independent hypothesis concerning their distribution. We consider this awkward to a higher degree, as it affects *local* quantities,

[†] Note that we have changed notation in comparison with our earlier papers, and that we are also using slightly different conventions—omitting in particular the parameter γ introduced in [1, 2] in favour of the interaction scale J . It should be no problem for the reader to translate results whenever desired.

whereas assumptions about the J_{ij} -distribution for $i \neq j$ contribute to the physics only via global *collective* effects less sensitive to details of the underlying assumptions.

3. Translationally invariant interactions

We now describe a modification of the above set-up in which the need for an independent hypothesis concerning the distribution of *local* variables is avoided, while at the same time having the additional benefit of providing an element of physics that had been missing in our original formulation.

To wit, the interpretation that we have given in support of the *ansatz* (2) is that the first harmonic contribution originates from a Born–von Karman expansion of the interaction energy about an (unknown) set of reference positions. Within such an interpretation, however, a bona fide interaction matrix ought to respect the principle of global translational invariance—a feature that had been missing in our original approach. One way of enforcing this principle is to require $\sum_j J_{ij} = 0$ for *all* i , entailing that the diagonal entries J_{ii} of the interaction matrix are *not independent* of the off-diagonal ones: $J_{ii} = -\sum_{j(\neq i)} J_{ij}$. In other words, translational invariance fixes the diagonal entries of the J -matrix solely in terms of true interaction contributions. It turns out that this rather slight modification has pronounced effects on the structure of the theory, which we have only just begun to explore.

Investigating the consequences of this idea quantitatively we choose a slightly different formulation, replacing (2) by

$$U_{\text{int}}(\{u_i\}) = \frac{1}{4} \sum_{i,j} J_{ij}(u_i - u_j)^2 + \sum_i G(u_i) . \quad (8)$$

It has the property that a global translation $u_i \rightarrow u_i + u$ (for all i) leaves the first contribution invariant, irrespective of the choice of the J_{ij} . As in [1, 2] we take the J_{ij} to be Gaussians, and we adhere to $G(u)$ being non-random and of the form (3), serving stabilizing purposes. The on-site potentials $G(u)$ do, of course, break translational invariance. However, we have also begun to look at variants in which the stabilizing contributions are themselves chosen in a translationally invariant form; results will be presented elsewhere.

Within a replica mean-field analysis, we now need additional order parameters beyond the polarization and the matrix of Edwards–Anderson order parameters to characterize the collective properties of the system, namely three replica correlations $q_{abc} = N^{-1} \sum_i \langle u_i^a u_i^b u_i^c \rangle$, and four replica correlations $q_{abcd} = N^{-1} \sum_i \langle u_i^a u_i^b u_i^c u_i^d \rangle$, the former, however, only for $a = b$, the latter either for $a = b = c = d$ or for $a = b$ and $c = d$.

So far we have studied the system only in a RS approximation. Details will be presented elsewhere. Here we only state the main results. In RS one assumes $p_a = p$, $q_{aa} = q_d$, and $q_{ab} = q$ for $a \neq b$ as before, and in addition $q_{aaa} = R_d$, $q_{aaaa} = Q_d$, and $q_{aab} = R$, $q_{aabb} = Q$ for $a \neq b$. It turns out that the four replica quantities cancel in RS expressions. The remaining order parameters are given as solutions of

$$\begin{aligned} p &= \langle \langle u \rangle \rangle_{y,z} & q_d &= \langle \langle u^2 \rangle \rangle_{y,z} & q &= \langle \langle u^2 \rangle \rangle_{y,z} \\ R_d &= \langle \langle u^3 \rangle \rangle_{y,z} & R &= \langle \langle u^2 \rangle \langle u \rangle \rangle_{y,z} . \end{aligned} \quad (9)$$

Here $\langle \langle \dots \rangle \rangle_{y,z}$ denotes an average over two uncorrelated standard Gaussians y and z , while $\langle \dots \rangle$ is a thermal average corresponding to the RS single-site potential:

$$U_{\text{eff}}(u) = -h_{\text{eff}}u + \frac{1}{2}k_{\text{eff}}u^2 + G(u) \quad (10)$$

with

$$h_{\text{eff}} = J_0 p - \frac{1}{2}J^2 C_R + J p y + J \sqrt{q - p^2} z \quad (11)$$

and

$$k_{\text{eff}} = J_0 - J^2 C + Jy \quad (12)$$

with $C = \beta(q_d - q)$ as before, and $C_R = \beta(R_d - R)$. Note that the structure of the theory is now no longer SK-like as in our original version. In particular, by enforcing translational invariance at the harmonic level of the Born–von Karman expansion, we now have two random variables characterizing the local effective potentials instead of one (on top of whatever local randomness one might wish to consider in the stabilizing on-site potentials $G(u)$).

4. Main results

We now turn to results. The main properties of the system are:

- (i) It exhibits a transition into a glassy phase with $q \neq 0$ at low temperatures (and small a_2).
- (ii) At sufficiently large J_0 , the transition is into a phase with macroscopic polarization $p \neq 0$.
- (iii) As in the original set-up, the transition into the glassy phase is continuous.
- (iv) In phases without macroscopic polarization we have $R_d = R \equiv 0$.
- (v) Though we have not yet performed the stability analysis, we expect RSB to occur at low temperatures (and small a_2).

With respect to the appearance of low-temperature anomalies, the following features deserve mention:

- (i) The potential energy landscape as represented by the ensemble of effective single-site potentials (10)–(12) exhibits DWPs and SWPs.
- (ii) The ensemble is now characterized by *two* random variables, a random effective field h_{eff} , and a randomly varying contribution k_{eff} to the harmonic force constant. As a consequence, DWPs will occur with a broad distribution of asymmetries and barrier heights, and both ‘soft’ and ‘hard’ SWPs will be observed.
- (iii) In phases with $p \neq 0$ the effective field h_{eff} and the harmonic force constants k_{eff} are correlated already in RS; we expect correlations between them to emerge irrespective of the value of the macroscopic polarization through RSB effects, much like in the original set-up [22].

At this point it is perhaps appropriate to connect our results with those of simulations performed to locate and characterize DWPs in Lennard-Jones systems [23]. DWPs observed in such systems can be parametrized (along the reaction path) by fourth-order polynomials of the form $U_{\text{DWP}}(u) = d_1 u + d_2 u^2 + d_4 u^4$ (or some equivalent form obtainable through shifts of coordinates). It is to be noted that such simulations will not have access to the full range of (d_1, d_2, d_4) triples owing to the non-linear constraint $d_2^3 + \frac{27}{8} d_1^2 d_4 \leq 0$ which characterizes DWPs, making it difficult to estimate the full distribution or to decide with confidence whether empirical correlations observed between the parameters are solely due to the non-linear constraint that defines the data set or of deeper origin. For the model considered in the present paper, we have the chance to compare such numerical results with analytic predictions, e.g. with the marginal distributions of d_1 and d_2 *conditioned on finding DWPs*. To compare with the theory, we have to identify $d_1 = -h_{\text{eff}}$, $d_2 = \frac{1}{2}(a_2 + k_{\text{eff}})$, and $d_4 = a_4/4!$. If we assume, for instance, a non-random d_4 and fix the energy scale by choosing $d_4 = 1$, and furthermore specialize to $J_0 = 0$, entailing $p = R_d = R = 0$, we obtain the following conditioned marginal probability densities:

$$p(d_1|\text{DWP}) = \frac{\text{constant}}{\sqrt{2\pi J^2 q}} \exp\left(-\frac{1}{2} \frac{d_1^2}{J^2 q}\right) \text{erfc}\left(\frac{3|d_1|^{2/3} + (a_2 - J^2 C)}{J\sqrt{2}}\right) \quad (13)$$

and

$$p(d_2|\text{DWP}) = \frac{\text{constant}}{\sqrt{2\pi J^2/4}} \exp\left(-\frac{1}{2} \frac{[d_2 - \frac{1}{2}(a_2 - J^2 C)]^2}{J^2/4}\right) \text{erf}\left(\frac{\sqrt{-\frac{8}{27}d_2^3}}{\sqrt{2J^2q}}\right). \quad (14)$$

These are compared with simulation results in figure 1. For d_1 the agreement is quite good, though not for d_2 . The remaining discrepancies are more probably due to RSB effects (which induce *a priori* correlations not related to the non-linear constraints) than to finite-size effects. Indeed, a major effect of RSB is expected to be a reduction of the value of C [2], which could already account for much of the observed discrepancy in the d_2 -distribution. It should be interesting to see how far our results might eventually carry to help in rationalizing the findings in Lennard-Jones systems.

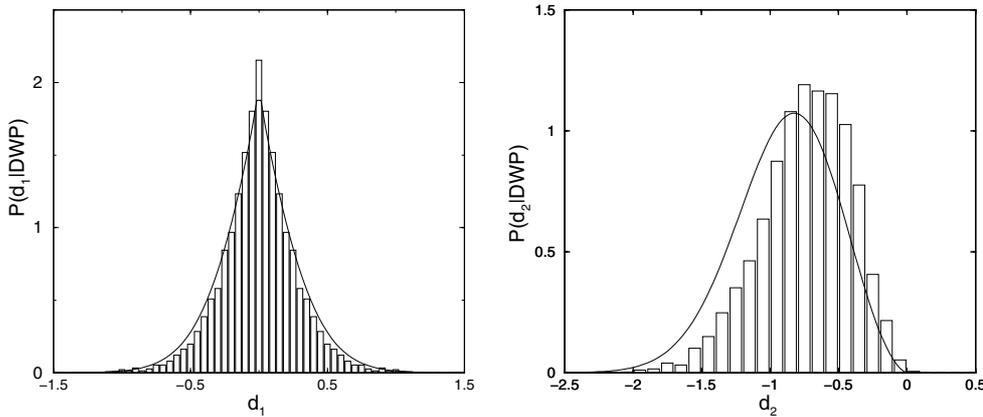


Figure 1. Marginal probability densities for the parameters d_1 (left) and d_2 (right) in an ensemble of DWPs generated by the interaction energy (10), with $J_0 = a_2 = 0$, $J = 1$, and $d_4 = a_4/4! = 1$. Full lines: the analytic prediction in the RS approximation. Simulation results were obtained for systems of size $N = 100$, using 1000 realizations so as to get reasonable statistics.

Continuing the list of results with a bearing on LTA:

- (iv) As DWPs in the glassy phase (and in polarized phases) occur with a broad spectrum of asymmetries (generated by the h_{eff} -distribution) they give rise to a broad spectrum of low-energy tunnelling excitations with a nearly constant DOS, and thereby to the universal low-temperature anomalies of specific heat and thermal conductivity.
- (v) As in the original set-up, higher-order excitations in DWPs and quasi-harmonic excitations in SWPs occur with a peaked DOS, producing a Bose peak.
- (vi) Because of the unbounded k_{eff} -distribution (Gaussian in the RS approximation), we no longer see a possibility of having amorphous phases *without* DWPs, unlike in the original set-up.
- (vii) Our previous analysis [2, 13], as to which low-temperature properties might be universal (those related to tunnelling excitations) and which not (those related to Bose peak phenomena), remains unaffected by the modifications of the present set-up.
- (viii) Last but not least, the proposal (8) embodies for the first time a *mechanism for generating a glassy potential energy landscape with an ensemble of SWPs and DWPs entirely in terms of interactions at the microscopic level via collective effects*, in which the distribution of both asymmetries and barrier heights of DWPs is amenable to analytic characterization and follows solely from assumptions about microscopic interactions. We regard this general

qualitative result as perhaps one of the most significant advances contained in the present contribution. Indeed, whereas there have always been plausible arguments concerning relevant features of the distribution of asymmetries of DWPs responsible for the LTA (broad, symmetric smooth, and hence nearly constant in the interesting energy range), such arguments have been missing for the distribution of barrier heights (or tunnelling matrix elements); within phenomenological modelling, it was always necessary to *guess* these distributions in a way that gets the main physics correct. Within our refined approach there is no longer any need for guesswork at this level; everything follows from assumptions about the interactions at the microscopic level via collective effects (and should—in view of the universality of glassy LTA—not critically depend on details of these assumptions). We are, of course, aware that this statement ought to be checked in greater detail than we have so far been able to do.

5. Concluding remarks

Whether the recent problems with the interpretation of some experiments at very low temperatures which we mentioned in our introduction are related to the fact that the above-mentioned guesses concerning tunnelling matrix elements (or concerning the absence of correlations between parameters characterizing local potential energy configurations) have not been entirely correct, or whether these problems point to more fundamental issues, we can at present not tell, as we have not yet been able to address these problems within the refined formulation of our spin-glass approach presented above.

The fact that the possibility for having phases without DWPs virtually disappears when global translational invariance is taken into account within a Born–von Karman expansion of the interaction energy casts some doubt on the interpretation of the low-internal-friction results of Liu *et al* [24] as being indicative of an amorphous system *without* low-energy tunnelling excitations. While the density of such excitations may indeed be unusually low in the a-Si samples of Liu *et al*, amorphous systems without DWPs appear to be extremely unlikely within our new perspective. This issue certainly deserves deeper investigation.

Among the interesting problems within reach of our approach we might mention in particular:

- (i) an analysis of collective quantum effects [19] via a fully fledged quantum statistical treatment of our spin-glass-type models [25],
- (ii) a deeper understanding of relations between LTA and the phenomenology at the glass transition,
- (iii) an investigation of potential energy landscapes which respects the three-dimensional nature of deviations from reference positions in view of possible consequences for magnetic field effects and Aharonov–Bohm phases in glasses [26], replacing (8) by

$$U_{\text{int}}(\{u_i^\mu\}) = \frac{1}{4} \sum_{i,j} \sum_{\mu,\nu} J_{ij}^{\mu\nu} (u_i^\mu - u_j^\mu)(u_i^\nu - u_j^\nu) + \sum_i G(\mathbf{u}_i)$$

in which μ and ν label the three Cartesian components of the \mathbf{u}_i ,

- (iv) the investigation of dynamic effects in the vicinity of the glass transition. In this respect finally note that—the glass transition in the present set-up being continuous—there is still need for improvements on the modelling side.

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