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## LETTER TO THE EDITOR

**A simple physical model of liquid–glass transition:  
intrinsic fluctuating interactions and random fields hidden  
in glass-forming liquids**

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**Abstract.** We propose that glass-forming liquids are intrinsically under the influences of both fluctuating interactions and random fields well-known in the field of spin systems. This is due to the frustration between the isotropic and anisotropic parts of effective intermolecular interactions. Our model indicates the existence of two key temperatures relevant to the glass transition, the density ordering point  $T_m^*$  and the Vogel–Fulcher temperature  $T_0$ . Between  $T_m^*$  and  $T_0$ , a system has features similar to the ‘Griffiths phase’, while below  $T_0$  it has those peculiar to the ‘spin-glass phase’. This picture naturally and universally explains vitrification behaviour from its strong to its fragile limit.

Although various features of the glass transition have been clarified recently [1–4], there has so far been no simple physical description of the glass transition covering its strong to its fragile limit. The theoretical approaches to this problem can be grouped into two types: (i) one focuses on the slowing down of the dynamics on approach to a glass-transition temperature  $T_g$ . The topological-confinement effects on molecular motion have been expressed in terms of a few different concepts, such as free volume [5], cooperative rearranging regions [6], and cage [7]. This stream leads to the development of the mode-coupling theory (MCT) [7]. However, the physical meaning of assumptions hidden in mode-coupling approximations is still not clear [8]. (ii) The other applies the knowledge of ‘spin glass’ [8–10], whose glassy behaviour is much more deeply understood than that of structural glass, and ‘frustrated systems’ [11, 12] to the problem of glass transition. This type of approach is attractive in the sense that it has a potential to provide us with a universal physical picture of random frustrated systems. These two approaches are essentially different from each other in that the former presupposes disorder, while the latter puts more emphasis on the ordering phenomena. It should be noted that neither of the above approaches provides us with any clear answer about what physical parameter controls the fragility of liquids [1, 2].

Before making any model of glass transition, we have to seriously consider a much more fundamental problem, namely, *why some molecules crystallize without vitrification while others can easily form glasses without crystallization*. Without considering ‘crystallization’, we cannot understand either a supercooled state, which is defined as a metastable state below a melting point, or vitrification phenomena, because the avoidance of crystallization is a prerequisite to them. Approach (i) regards the glass transition as a purely dynamic transition. Thus, it usually neglects even the fact that a glass-forming system has the ability of crystallization. Approach (ii), on the other hand, fails to give a clear molecular-level

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explanation of the physical origin of the similarity between spin glass and structural glass, although it recognizes the importance of topological frustration [8, 9, 10]. Thus, the wide gap between them has not been filled yet. Hence, neither approach can clearly explain why slow dynamics start to appear when a liquid enters into a metastable supercooled state upon cooling, namely, when a liquid is cooled below the melting point of its crystal.

The main aim of this letter is to clarify the physical origin of frustration in glass-forming liquids that apparently have no intrinsic quenched disorder, and to establish a direct connection between structural glass and spin glass on the basis of molecular-level consideration. In contrast to the common sense that density  $\rho$  is the only order parameter required for the physical description of liquids, we propose that the introduction is necessary of a new order parameter (bond parameter) representing a locally favoured structure in liquids. This local bond formation causes ‘fluctuations in the intermolecular interactions’ and ‘symmetry-breaking random fields’ in much the same way as in spin systems [8, 14]. On the basis of the knowledge of spin systems [14], we propose a universal picture of the glass transition. To our knowledge, this is the first approach to the problem of the liquid–glass transition directly focusing on ‘crystallization (or density ordering)’.

First we focus on the effective attractive interaction potential between a molecule and its neighbouring molecules. It is generally given by the form  $V(r, \Omega) = \bar{V}(r) + \Delta V(r, \Omega)$ , where  $r$  is the distance from the centre of mass of the molecule and  $\Omega$  expresses the orientation.  $\bar{V}$  represents the isotropic part of the interaction and  $\Delta V$  its anisotropic part. This anisotropy can lead to a locally favoured structure made of a molecule and its neighbouring  $n$  molecules, which is not consistent with the crystallographic symmetry. Thus, there can exist competing interactions in any liquids. Even for spherical-particle systems, it is known that an ‘icosahedral structure’ is locally favoured, which is separated from alternative arrangements such as bcc and fcc by high potential energy barriers ( $\sim n\Delta V$ ,  $n = 12$ ). Its importance in the glass problem was widely recognized by many researchers [9–12, 15–17]. This local bond ordering is incompatible with any long-range order, so that it plays the role of a random disordering field against crystallization: it favours vitrification. This competition between density and bond ordering results from two conflicting requirements: (i) to minimize the distance between nearest-neighbour molecules; and (ii) to maximize the number of surrounding molecules. In strong liquids, on the other hand, the locally favoured symmetry is mainly selected by specific anisotropic interactions between molecules [18] such as hydrogen and covalent bonding, which often lead to a ‘tetrahedral structure’. For a general physical description of real liquids, thus, we need a new order parameter  $S$  to describe the presence of such locally preferred arrangement of liquid molecules:  $\rho$  and  $S$  are the minimal order parameters required for the physical description of the above complex features of many-body interactions. Although  $S$  should have a tensorial character that plays important roles in the selection of crystallographic symmetry and its rotational dynamics, we can treat it as a scalar order parameter  $S$  when we consider the phase behaviour. The relevance of such an approximation is well established in the field of liquid crystals [19]. Without losing generality, therefore, the bond parameter  $S$  can be defined as the ‘local number density of locally favoured structures’:  $S(\mathbf{r}) = \sum_i \delta(\mathbf{r} - \mathbf{r}_i)$ , where  $\mathbf{r}_i$  is the position vector of a locally favoured structure (number  $i$ ) and  $\sum_i$  is the sum about  $i$  over a unit volume. Note that the spatial distribution of locally favoured structures ( $\mathbf{r}_i$ ) is random due to the nature of bond formation [20]. The spatially averaged value of  $S$  is given by  $\bar{S} = S_0 \exp(\beta n \Delta V)$ , where  $\beta = 1/k_B T$  ( $k_B$  is the Boltzmann constant and  $T$  is the temperature), since a locally favoured structure, which is stabilized by  $n$  bonds, is in a lower energy state than the other part of the liquid by  $n\Delta V$ . When the dependence of  $\Delta V$  on  $\Omega$  is not consistent with the crystallographic symmetry,  $S$  decreases the local density,

lowers the crystallization temperature and thus locally disturbs crystallization. Thus, each molecule itself internally has the cause of disorder and random fields against the density ordering. We believe that this frustration between  $\rho$  and  $S$  is the physical factor determining how easily molecules can form a glass without crystallization. Note that the strength of disorder effects increases with  $\Delta V$ , and so the stronger glass suffers from the stronger spin-glass (SG) effects.

The above consideration leads to the following two-order-parameter description of liquids. In contrast to the common sense that a liquid state can be described by a single order parameter,  $\rho$ , we need a new order parameter  $S$  to express a tendency of local bond ordering since in any liquids molecules locally favour a certain symmetry, which is not necessarily consistent with a crystallographic symmetry favoured by density ordering. This competition between the two order parameters,  $\rho$  and  $S$ , causes frustration, which we believe is the major origin of vitrification. This physical picture can be naturally modelled by a Ginzburg–Landau-type model with the couplings between  $\rho$  and  $S$ , which represent disorder effects of  $S$  on the ordering of  $\rho$ . Density fluctuations  $\delta\rho$  in the liquid phase indicating the instability toward the solid phase have a maximum at nonzero wavenumber  $q_0$ , whose essential feature is well characterized by the following bare static structure factor:  $S_0(q) = 2k_B T \chi_0(q) = k_B T / [\tau + K(q - q_0)^2]$ . The phenomenological free energy which predicts the above  $S_0(q)$  is given by [19]

$$\beta H_\rho = \int d\mathbf{r} d\mathbf{r}' \delta\rho(\mathbf{r}) \chi_0^{-1}(\mathbf{r} - \mathbf{r}') \delta\rho(\mathbf{r}') - \frac{f}{3} \int d\mathbf{r} \delta\rho(\mathbf{r})^3 + \frac{g}{4} \int d\mathbf{r} \delta\rho(\mathbf{r})^4 \quad (1)$$

where  $\tau = a_2(T - T_\rho^*)$  and  $T_\rho^*$  is the temperature of the mean-field limit of stability of the liquid phase. This Hamiltonian implies that the ‘density order parameter favours spherical symmetry’. By including the coupling between  $\delta\rho$  and  $S$  into the above standard theory of a liquid–solid transition, we obtain the following Hamiltonian which we believe is relevant to the glass transition:

$$\beta H_{GT} = \beta H_\rho + \int d\mathbf{r} \left( -c_1 \delta\rho(\mathbf{r}) S(\mathbf{r}) - \frac{c_2}{2} \delta\rho(\mathbf{r})^2 S(\mathbf{r}) \right). \quad (2)$$

In the above, the coupling between  $\rho$  and  $S$  is introduced through the coupling constants  $c_i$ . For the ‘negative’ coupling ( $c_i < 0$ ), the formation of active bonds (or  $S$ ) leads to a decrease of local density and also to the decrease of the ordering temperature. For the ‘positive’ coupling ( $c_i > 0$ ), on the other hand, the formation of active bonds (or  $S$ ) leads to an increase in local density and to an increase of the ordering temperature, so molecules should never form a glass and just crystallize. *The type (sign) of coupling between  $\rho$  and  $S$  gives a simple criterion on whether molecules crystallize without vitrification or can easily form a glass.* Hereafter we consider the case of  $c_i < 0$ , since vitrification can occur only for this case.

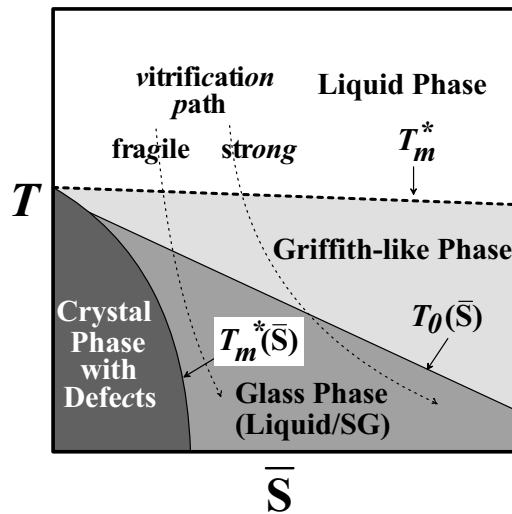
The dynamics of  $\delta\rho$  can be described by [10]

$$\frac{\partial \delta\rho(\mathbf{r}, t)}{\partial t} = \Gamma_0 \nabla^2 \frac{\delta(\beta H_{GT})}{\delta(\delta\rho(\mathbf{r}, t))} + \zeta(\mathbf{r}, t) \quad (3)$$

where  $\zeta$  is the noise term and  $\Gamma_0$  is a bare kinetic coefficient. Although  $S$  is not a quenched variable, the much slower dynamics of  $S$  than  $\rho$  guarantees the quasi-quenched nature of  $S$ : The lifetime of locally favoured structures is longer than the characteristic time of density fluctuations by at least a factor of  $R \sim \exp(\beta n \Delta V)$ . More importantly, recent theoretical studies indicate that even a frustrated random system without quenched disorder has essentially the same features as that with quenched disorder [8]. These facts allow us

to regard  $S$  as a quenched variable as long as we consider disorder effects of  $S$  on density ordering. Thus, we treat  $S$  as a quenched variable hereafter.

In relation to the above, we briefly consider the stability of a metastable supercooled state. First we define  $T_m^*(0) = T_m^*$  and  $T_m^*(\bar{S})$  respectively as the density-ordering temperature (a crystallization or melting temperature) without and with the effects of random disorder ( $\bar{S}$  is the disorder strength). The former is the melting temperature of a real defect-free crystal formed as a result of density ordering, while the latter is that of a hypothetical crystal with quenched disorder (or local bond ordering), which may not exist in reality. The melting temperature of this hypothetical crystal with defects,  $T_m^*(\bar{S})$ , rapidly decreases with an increase in defect density, or  $\bar{S}$  (see figure 1), which is known as ‘dilution effects’ in spin systems. For real crystallization, which requires the annealing of the random nature of  $S$ , on the other hand, a system has to overcome a large energy barrier corresponding to the break-up or deformation of all the active bonds in a critical nucleus of crystal, which costs an energy of the order of  $\Delta E \sim n\Delta V\bar{S}v_n$ , where  $v_n$  is the volume of a critical nucleus. This naturally explains why a metastable supercooled state is so stable in glass-forming liquids. Crystallization at  $T_m^*$  can thus be kinetically avoided by this extra energetic barrier for nucleation,  $\Delta E$ , for a sufficient cooling rate and thus vitrification can be induced, as described below. In the metastable branch of a supercooled liquid, therefore, the random distribution of locally favoured structures is not altered [20] by density ordering. Provided that crystallization is kinetically prohibited, we can regard a supercooled state as the quasi-equilibrium thermodynamic state. Thus, we consider a problem of vitrification on the basis of this quasi-equilibrium assumption.



**Figure 1.** Schematic phase diagram of the liquid–glass transition.  $\bar{S}$  is a measure of disorder strength against density ordering. SG stands for spin-glass phase. Crystallization occurs around  $T_m^*$  if it is not kinetically avoided.

Here we point out the similarity of the above Hamiltonian  $H_{GT}$  (see equation (2)) assuming a quenched nature of  $S$  and that of a spin system under fluctuating interactions and random fields. The Ginzburg–Landau-type Hamiltonian under random transition

temperatures  $\delta\tau(\mathbf{r})$ ,  $H_{QD}$ , and that under random fields  $h(\mathbf{r})$ ,  $H_{RF}$ , are [14]

$$\beta H_{QD} = \int d\mathbf{r} \left[ \frac{1}{2} K (\nabla\phi)^2 + \frac{1}{2} (\tau - \delta\tau(\mathbf{r})) \phi^2 + \frac{1}{4} g \phi^4 \right]$$

$$\beta H_{RF} = \int d\mathbf{r} \left[ \frac{1}{2} K (\nabla\phi)^2 + \frac{1}{2} \tau \phi^2 + \frac{1}{4} g \phi^4 + h(\mathbf{r}) \phi \right]$$

where  $\phi(\mathbf{r})$  is the order parameter. Our Hamiltonian  $H_{GT}$  has both features of  $H_{QD}$  and  $H_{RF}$  at the same time. By setting  $\phi(\mathbf{r}) = \delta\rho(\mathbf{r})$ ,  $\delta\tau(\mathbf{r}) = c_2 S(\mathbf{r})$  and  $h(\mathbf{r}) = -c_1 S(\mathbf{r})$ ,  $H_{GT}$  can be directly correlated to  $H_{QD}$  and  $H_{RF}$ , after coarse graining the high  $q$  mode around  $q_0$ . Thus, *the essential effects of random disorder on density ordering in our system should be the same as those in spin systems* [8, 14]. The quasi-quenched nature of  $S$  allows us to consider the problem in terms of complex energy landscape, without solving the dynamic equations.

On the basis of our knowledge of a spin glass [8, 14], we draw a simple physical picture of glass transition (see figure 1). Above  $T_m^*$ , the system behaves just as an ordinary liquid and the structural relaxation time  $\tau_\alpha$  obeys a simple Arrhenius law,  $\tau_\alpha = \tau_\alpha^\infty \exp(\Delta G/k_B T)$ , where  $\Delta G$  is the activation energy and an increasing function of  $\Delta V$ , and the relaxation function  $\Phi(t)$  is exponential:  $\Phi(t) = \Phi(0) \exp(-t/\tau_\alpha)$ . Below  $T_m^*$ , the saddle-point equation  $\delta H_{GT}/\delta(\delta\rho) = 0$ , namely  $-K \nabla^2 \delta\rho(\mathbf{r}) + [\tau - c_2 S(\mathbf{r})] \delta\rho(\mathbf{r}) + g \delta\rho(\mathbf{r})^3 - c_2 S(\mathbf{r}) = 0$ , starts to have local minimum solutions around the high-density side of the liquid free-energy minimum [21]. This means there exist a macroscopic number of spatial ‘islands’ having a higher density than the liquid, below  $T_m^*$ . This situation is similar to the spin glass where the phase space is factorized into a hierarchy of ‘valleys’, or pure states of local minima separated by macroscopic barriers. Thus, slow relaxations overcoming barriers separated by different valleys are expected. This phase existing between  $T_m^*$  and  $T_m^*(\bar{S})$ , which is characterized by the existence of numerous metastable states separated by finite barriers, is similar to the ‘Griffiths phase’ [22, 23] known in spin systems.

This ‘Griffiths-like phase’ can be characterized as follows [14, 22, 23]. In the temperature interval  $T_m^*(\bar{S}) < T < T_m^*$ ,  $\Phi(t)$  is described by the stretched exponential [23, 24]:  $\Phi(t) = \Phi(0) \exp[-(t/\tau_\alpha)^{\beta_K}]$ , instead of the usual exponential relaxation, as it should be in the ordinary liquid phase (the paramagnetic phase in spin systems) [25]. In spin systems [26], it is claimed that the stretched exponential parameter  $\beta_K$  is the temperature-dependent exponent, as it is a finite value ( $< 1$ ) at  $T = T_m^*(\bar{S})$ , and it increases monotonically up to  $\beta_K = 1$  at  $T_m^*$ . This is consistent with what has been observed in structural glasses [3, 4, 25]. Further, the strong coupling between ‘islands’ probably leads to the existence of the SG-like phase below  $T_{SG}$ , in which the numerous disorder-dependent local minima are probably separated by macroscopic (or infinite) energy barriers [14]. We assign this phase transition at  $T_{SG}$ , from the Griffiths-like phase to the SG-like phase, to the Vogel–Fulcher temperature  $T_0$ , where the relaxation time diverges due to the infinite barriers and the ergodic to non-ergodic transition takes place. However, this point is never reached in real experiments because the large barrier heights near  $T_0$  cause extremely slow relaxations even above  $T_0$ . Then, the glass-transition temperature  $T_g$  can be defined as a temperature where the metastable ‘islands’ having sufficiently high energetic barriers do percolate. Thus,  $T_m^* > T_g > T_0 > T_m^*(\bar{S})$ .

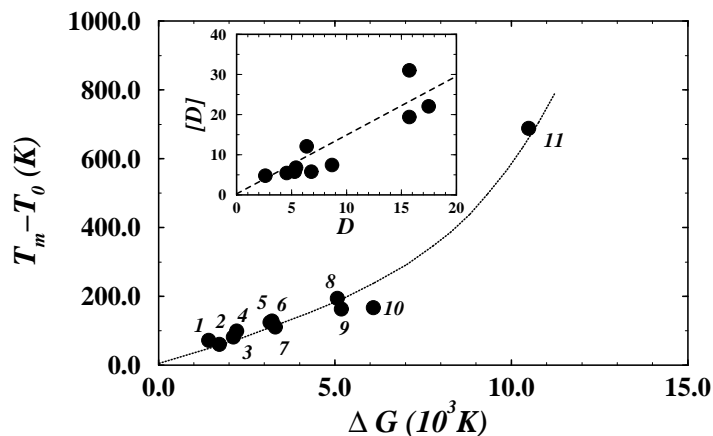
Figure 1 shows a phase diagram of glass transition on the basis of the above physical picture. Here it should be noted that for the liquid–glass transition there can be two types of the origins of disorder whose temperature dependences are crucially different from each other: (i) anisotropic interactions that are not consistent with the crystallographic symmetry, in molecular glass formers; and (ii) quenched disorder in structures of particle or molecules,

e.g. the polydispersity of colloid particle sizes and the disorder in the stereoregularity, tacticity, and chemical structures of polymers. For case (i) the strength of disorder and random fields ( $\bar{S}$ ) is strongly dependent upon  $T$ , while for case (ii) the strength of the disorder is independent of  $T$ . The temperature-cooling paths drawn in figure 1 correspond to case (i).

Here we consider the non-Arrhenius behaviour below  $T_m^*$  in more detail. We assume the divergence of the barrier height  $E_{barrier}$  between ‘islands’ is given by  $E_{barrier} = BT/(T - T_0)$ . This gives us the well-known Vogel–Fulcher (VF) law. Here we do not repeat the argument deriving this relation (see e.g. [9, 10, 14, 24, 27] for details). Instead, we stress that the effective barrier dominating the structural relaxation  $E_{barrier}^{eff}$  should be given by (i)  $E_{barrier}^{eff} = \Delta G$  above  $T_m^*$  and (ii)  $E_{barrier}^{eff} = \Delta G + BT/(T - T_0)$  below  $T_m^*$ . This can be expressed analytically by the following modified VF law:

$$\tau_\alpha = \tau_\alpha^\infty \exp(\Delta G/k_B T) \exp[f(T)B/k_B(T - T_0)]. \quad (4)$$

Here  $f(T)$  is the Fermi-distribution-like function  $f(T) = 1/\{\exp[\gamma(T - T_m^*)] + 1\}$ , where  $\gamma$  is a positive constant that controls the sharpness of the transition at  $T_m^*$ . This modified VF law can naturally describe the crossover from the Arrhenius to non-Arrhenius behaviour at  $T_m^*$  and the divergence at  $T_0$ . The fitting of the traditional VF law to data in a low temperature range often produces an unrealistic attempt frequency  $\tau_\alpha^\infty$  (see e.g. [28]). This problem can be removed by using equation (4). From the condition of the disappearance of the non-Arrhenius behaviour above  $T_m^*$ , further, we obtain the relation  $B \sim \Delta G(T_m^* - T_0)/T_m^*$ . The fragility parameter [ $D$ ] estimated by this relation with  $[D] = B/T_0$  is well correlated with  $D$  determined experimentally, as shown in the inset of figure 2. This provides us with a clear physical reasoning on the Angell plot [13], including the correlation between bond strength  $S$  and the strong nature of liquids.



**Figure 2.** Relation between  $\Delta G$  and  $T_m - T_0$ . The data are taken from [12]. 1: *n*-butyl benzene; 2: isopropyl benzene; 3: propyl carbonate; 4: *n*-propanol; 5: *o*-terphenyl; 6: salol; 7: dibutyl phthalate; 8: *s*-triphthyl benzene; 9: glycerol; 10:  $\alpha$ -phenyl-cresol; 11: boron oxide. The curve is to guide the eye. The inset shows the relation between  $[D]$  and  $D$  (see the text for their definitions).

Finally, we check the other main predictions of our model: (i) there occurs a dynamic transition from the non-cooperative (Arrhenius-type) to cooperative regime (Vogel–Fulcher-type behaviour) at  $T_m^*$  (near  $T_m$ ); (ii) the disorder strength  $\bar{S}$ , which can be correlated

with the activation energy  $\Delta G$  of the  $\alpha$  relaxation above  $T_m^*$ , determines the temperature interval between  $T_m^*$  and  $T_0$ ; (iii) a stronger glass suffers from stronger disorder effects; (iv) an increase in pressure increases  $\bar{\rho}$ , but decreases  $\bar{S}$  for  $c_i < 0$ , simply because a locally favoured structure having a greater volume is destroyed by applying pressure. This weakens the random disorder effects, which makes the system more fragile. Prediction (i) is quite natural and has already been recognized by many researchers (see e.g. [28]), although there has so far been no physical reasoning on it. We stress that none of previous models can explain this fact, since no models of glass transition have so far focused on crystallization phenomena themselves. This prediction is quite specific in the sense that  $T_m^*$  is a measurable quantity, while  $T_c$  in MCT and  $T^*$  in the theory of Kivelson *et al* are not. Prediction (ii), on the other hand, can be confirmed by figure 2, which indicates the strong correlation between  $\Delta G$  (or  $\Delta V$ ) and  $T_m - T_0$ . Prediction (iii) is consistent with: (a) the fact that the change in specific heat across  $T_g$  is weaker for a stronger glass (see e.g. figure 4 of [1]); (b) the recent finding of Sokolov *et al* [29] on the stronger phonon scattering in stronger liquids; (c) a larger interval between  $T_g$  and  $T_0$ ; and (d) the resulting larger  $\beta_K$  at  $T_g$  for a stronger glass (see e.g. [3]). Prediction (iv) is also quite consistent with the experimental findings of Cook *et al* [1, 30] that strong liquids become more fragile under a high pressure. This cannot be explained by a theory like MCT that is described by a single order parameter (density), since a plot of viscosity versus density should be universal in such models. The relevance of our two-order-parameter (spin-glass) model to the glass transition is strongly supported by the validity of these predictions and also by the experimental results suggesting the similarity between structural glass and spin glass [31].

It is worth noting here the relation between our model and the model of Kivelson *et al* [11, 12], since both focus on ‘frustration’. The most crucial difference is that we take a crystalline state (the ordering temperature  $T_m^*$ ) as the reference state, while they take a ‘postulated’ quasicrystal of a locally favoured structure (the ordering temperature  $T^*$ ), which is prohibited in a real system due to frustration. In other words, they presuppose the avoidance of crystallization by hand. This leads to essential differences in physics, although both temperatures are claimed to be located near  $T_m$ . The correlation of  $T_m^*$  to  $T_m$  is quite natural for our case, while there seems to be no such justification for their case since we cannot expect any correlation between the hidden ordering point of a frustrated quasicrystal and that of a real crystal, which have essentially different symmetries.

In summary, we propose a simple physical picture of glass transition on the basis of ‘the two-order-parameter description of liquids’ [32], which connects structural glass and spin glass in a natural way. In our view, vitrification is a result of the competition between density ordering and hidden local bond ordering. Our study indicates that frustrated systems such as spin glass and structural glass have the universal phase and dynamic behaviour characterized by a complex energy landscape peculiar to ‘Griffiths-like’ and ‘spin-glass’ phases. Here it should be noted that the fragile limit ( $S \rightarrow 0$ ) with  $c_2 = 0$  of the dynamic version of our model is mathematically identical to the pioneering theory of Kirkpatrick and Thirumalai [10], which further has dynamic features similar to MCT [7] (see the discussions in [10]). Thus, our model may give a clue to the problem of the existence of hidden disorder in mode-coupling equations pointed out by Bouchaud *et al* [8]. Finally, we stress that our model can provide us with *a universal description of the glass transition covering its strong to fragile limit*. The strong nature of liquids increases with an increase in the disorder strength against density ordering ( $c_1 S$  and  $c_2 S$ ).

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