Correlated random fields in dielectric and spin glasses

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Abstract – Both orientational glasses and dipolar glasses possess an intrinsic random field, coming from the volume difference between impurity and host ions. We show this suppresses the glass transition, causing instead a crossover to the low-\(T\) phase. Moreover the random field is correlated with the inter-impurity interactions, and has a broad distribution. This leads to a peculiar variant of the Imry-Ma mechanism, with “domains” of impurities oriented by a few frozen pairs. These domains are small; predictions of domain size are given for specific systems, and their possible experimental verification is outlined. In magnetic glasses in zero field the glass transition survives, because the random fields are disallowed by time-reversal symmetry; applying a magnetic field then generates random fields, and suppresses the spin glass transition.

Most real solids are either amorphous or disordered crystals, and many of these show some sort of “glassy” behavior at low temperatures [1]. These glasses have been categorized into different types, including spin [2], structural [3], electron [4], orientational [5], and dipolar glasses [6]. All of these show aging, rejuvenation and memory effects. However, thermodynamically there is a marked difference between systems like spin glasses (SGs), which show a clear phase transition between the paramagnetic and SG phases (with diverging length scale and consequent diverging nonlinear susceptibility [7]), and systems like orientational glasses (OGs) or electric dipole glasses (DG) where the transition to the low-\(T\) phase is smeared [6,8,9].

At first glance, this difference seems paradoxical—in OGs, the rotational degree of freedom behaves like an Ising pseudospin 1/2, similar to that in DGs; and the electric- and strain-mediated interactions between these pseudospins have a similar form to those between the real spins in dipolar SGs. The quenched randomness then implies that OGs and DGs should show the same critical behavior as dipolar SGs, that of the short range Edwards-Anderson model [10].

In the present paper we argue that the full answer to this question is found in an analysis of the effective Hamiltonians for these systems—including the form of the random fields (RFs) that are generated—and in the constraints imposed by symmetry. These RFs are broadly distributed, and correlated with the interactions; we find this leads to a peculiar disordering mechanism, intermediate between simple alignment along the RFs and Imry-Ma [11] behavior.

Orientational and dipolar glasses. – For typical OGs like KBr:CN, various approaches have been used to describe the interactions [12–15]. If one adds to the standard strain-mediated interaction [14,16] another term reflecting the volume difference between host and impurity ions, an effective random field (RF) is found to be generated by theorientational impurities themselves. The OG degrees of freedom can be treated formally in the same way as the pseudospin degrees of freedom in DG systems like Li:KCl, allowing one to analyse the interactions of both systems on the same footing [15].

One then finds [15] that over a wide range of temperatures extending up to the putative glass transition temperature \(T_g\), both OGs and DGs are described by the pseudospin effective Hamiltonian

\[ H_{\text{eff}} = \sum_{ij} U_{ij}^{zz} \hat{\sigma}_i^z \hat{\sigma}_j^z + \sum_j b_j \hat{\sigma}_j^z. \]  

(1)

We emphasize that both the Ising interaction \(U_{ij}^{zz}\) and the RFs \(\{b_j\}\) are generated by the same defect-phonon
interaction
\[
V_{dp} = -\sum_{\alpha,\beta} (\eta_0 \delta_{\alpha\beta} + \gamma_{ij} \sigma_j^\alpha ) \frac{\partial X_{j\alpha}}{\partial x_{j\beta}}
\]
expanded to second order in perturbation. Here \(X_{j\alpha}\) represents the displacement operator at point \(j\) in direction \(\alpha\). One finds \([15]\) \(U_{ij}^{zz} = g_{zz} \tau_{ij}^{zz}/R_{ij}^3\), and \(b_j \equiv \sum_i U_{ij}^{zz} = g_{zz} \sum_i \tau_{ij}^{zz}/R_{ij}^3\) with \(R_{ij} = r_i - r_j\), and where \(g_{zz} \sim \gamma_i^2/\rho_c\), and \(g_{zz} \sim (\gamma_i^2/\rho_c^2)\). Here \(\rho\) is the density, \(c_{\alpha}\) \((c_1)\) is an average (longitudinal) sound velocity, and \(\tau_{ij}^{zz}, \tau_{ij}^{0z} \sim O(1)\) are complicated angular averages over the tensor components of the interaction.

The typical parameters of the Hamiltonian \((1)\) are therefore given by
\[
\bar{U} \approx \frac{\gamma^2}{\rho c_0} n, \quad \tilde{b} \approx \frac{\eta_0 \gamma_0}{\rho c_0} n
\]
and we note that the ratio \(\bar{U}/\tilde{b}\) is independent of \(n\).

In principle, one should add to the Hamiltonian \((1)\) a tunneling term \(\sum_i \Delta_i \sigma_i^3\), which is crucial for the dynamics of the system. However, we are interested in the case where interactions dominate, i.e., \(\bar{U} > \Delta\); the tunneling term is then irrelevant when considering the phase diagram. For concentration \(n = 10^{-3}\), typical values are \(\bar{U} \sim 1.5\,K\) and \(\tilde{b}\) somewhat smaller. Typically we assume \(n < 1\) in what follows, but we will also compare with larger \(n\).

**Correlated random fields.** – Consider now the RFs \(\{b_i\}\). These were previously argued to lead to a glassy state \([13]\), or to a state intermediate between a true spin glass and a pure random-field system \([17]\). Here we argue that actually the RFs destroy long-range glassy order in the low-\(T\) phase \([18]\), which is then reached by a crossover and not a phase transition. Furthermore, since the RFs originate from the impurity lattice interactions themselves, they are correlated with the interactions (because \(U_{ij}^{0z}\) is correlated with \(U_{ij}^{zz}\)) and have a broad distribution. When defects/impurities occupy nearest-neighbour sites, the field of one on the other is \(b_i \approx \eta_0 \gamma_0 / (\rho c_0^2) > b\).

Because \(U_{ij} \propto 1/R^3\) and is random, \((U_{ij}^{zz}) \propto 1/R^6\) and is effectively short range \([10,18]\). The situation is thus similar to that analysed by Imry-Ma \([11]\), and its generalisation to glasses by Fisher and Huse \([18]\). However there is an important difference—we find that in systems like dilute OGs, where the RFs are broadly distributed and are correlated with the interactions, one gets qualitatively different results from systems where the RFs are normally distributed. The largest RF, occurring in the event when defects/impurities occupy nearest-neighbour sites, is \(b_0\). Since \(n < 1\), such events are rare; however the average RF \(b_{\bar{n}}\) \((the \ standard \ deviation \ of \ the \ distribution)\) is dominated by these rare events, and is given by \(b_{\bar{n}} \approx \eta_0 \gamma_0 / (\rho c_0^2) > b\) \((see \ ref. \ [19], \ noting \ the \ trivial \ relations \ between \ high \ and \ low \ impurity \ densities)\). Now, since \(b_0 \ll U_0\) \((where \ U_0 \ is \ the \ interaction \ between \ nearest-neighbour \ defects/impurities)\), two regimes are possible, viz.: a) \(b_0 \ll U_0n\) and b) \(U_0n \ll b_0 \ll U_0\).

Regime a) is simple. All RFs are smaller than the typical interaction. One can then follow the standard application of the Imry-Ma argument \([11]\) to SGs \([18]\). The energy cost to flip a domain is related to the typical interaction, and therefore is \(\sim \bar{U}L^6\), where \(L\) is the domain size, and the stiffness exponent \(\theta \approx 0.2\) in 3D \([18,20]\). However, the effective RF is given by \(b_{av} = b_0\sqrt{n} = \sqrt{b_0}\). Thus, the number of spins in a domain \(N_0\), and the related dimensionless correlation length \(\xi \equiv \xi n^{1/3} \sim N_0^{1/3}\) are both \(n\)-dependent, the latter being given by
\[
\xi \approx \left( \frac{U_{0n}}{b_0\sqrt{n}} \right)^{1/3} \approx n^{0.39} \left( \frac{\bar{U}}{b} \right)^{0.78}
\]
In comparison to Gaussian-distributed RFs, the domain size here is smaller by a factor \(n^{0.39}\).

Regime b) \((which \ usually \ applies \ in \ dilute \ dipolar \ glasses)\) is more complex, because the spread of RFs is larger than the typical interactions, so Imry-Ma arguments cannot be used directly. Consider a pair of nearest-neighbour impurities/defects \(l, l'\) \((3\)-impurity correlations can be neglected when \(n \ll 1\). Since \(U_0 \gg \bar{U}\), the two impurities will mutually order, i.e., lock together. If this locking is antiferromagnetic, then the net RF on the pair comes only from distant impurities —the internally generated RF \(U_{0l}^{0z} - U_{0l'}^{0z} = 0\), since \(U_{ij}^{0z} = U_{ij}^{0z}\) for any pair \(i, j\). However, for ferromagnetic ordering the internally generated RF is \(\sim b_0\), and since \(U_0n \ll b_0\), this pair will be oriented along this internal RF, and hardly influenced by the interactions with other impurities, which have strength \(\sim \bar{U} = U_0n\)! In fact, this is the case for all ferromagnetically aligned impurity pairs satisfying \(b_0/R_{ij}^3 > \bar{U}\). In a sample of \(N\) impurities there are \(\approx N b_0/U_0\) such “frozen pairs”. Consider now a sample containing \(N\) impurities/defects. The typical RF resulting from the frozen pairs is then \(\sim \sqrt{b_0N/U_0U_{0n}}\), while that from all other impurities is only \(\sim \sqrt{N}b_0\). Thus despite their scarcity, the RF from the frozen pairs dominates the total RF. Using now the standard argument which compares the energy gain from the RF and the interaction energy cost at domain boundaries, we find for a dilute DG in regime b) a dimensionless correlation length given by
\[
\xi \approx \left( \frac{\bar{U}}{\sqrt{\bar{U}b}} \right)^{1/3} \approx \left( \frac{\bar{U}}{b} \right)^{0.39}
\]
Thus, in this regime the correlation length is determined neither by the typical RF \(b_0\), nor by the average RF \(b_0\sqrt{n}\), but by the algebraic mean of \(\bar{U}\) and \(b\). This is because the relaxation of the RFs occurs in two stages. The rare pairs with \(b_i > \bar{U}\) relax locally to their frozen state. The majority of the spins then order via the Imry-Ma mechanism, albeit with RFs dominated by their interaction with the rare frozen pairs. Because \(\bar{U}/b\) is scale-invariant \(N_d \sim (\bar{U}/b)^{1.17}\), independent of \(n\). The low power in the exponent is dictated by the unique mechanism above, and results in “quasi-domains”, which in practise
contain very few defects/impurities (see the examples below). Moreover, in each such quasi-domain there are only \( N / b \approx 1 \) frozen pairs. In this limit one cannot of course assume that the random fields behave as in the large \( N \) limit; however, the mechanism described above gives an upper limit to the size of the domains, since it minimizes the effect of the RFs. A lower limit can be given by the assumption that the total RF from the frozen pairs is maximized, leading to \( N_d \sim (U / b) \). In practice these two limits are indistinguishable, suggesting that domains are formed in such a way that interactions with frozen pairs are maximized.

Thus, in regime b) we arrive at a picture of very small quasi-domains or “clusters” of \( N_d \) pseudospins, of size \( \xi \), each containing \( \sim O(1) \) frozen pseudospin, whose field then orients the rest of them. For \( n \approx 1 \) (regime a)) we find \( N_d \approx (U / b)^{1/2} \), giving large clusters, but on strong dilution this crosses over to the much smaller \( N_d \approx (U / b)^{1/2} \). We note that for all dilutions, the finite \( \xi \) gives a finite nonlinear susceptibility \( \chi_3 \propto \xi^{4-n} \), marking a crossover rather than a transition to the glassy state \([21,22]\), in agreement with experiments \([6]\). However, the decrease in \( N_d \) with \( n \) means that the peak in the nonlinear susceptibility should be much smaller for dilute OGS and DGs, in comparison with similar systems at high concentrations. This is a central prediction of this letter.

In real systems, the ratio \( U / b \) varies widely between different OGS and DGs, and it is useful to look at some experimental examples. Consider first OH- impurities in KCl, where the electric and elastic impurity-impurity interactions are comparable, and \( U / b \approx 3 \) (see ref. \([23]\)). In ref. \([9]\) it was argued, based on the behavior of the nonlinear dielectric permittivity of dilute KCl:OH, that there is no transition to a glass phase, but rather to a state analogous to a superparamagnet, with roughly 10 impurities per domain. The scaling approach we use here, embodied in eq. (5), is strictly applicable only for \( \xi \gg 1 \), and even then only gives an order of magnitude. Thus our result here is consistent with the above experimental picture of small “superparamagnetic” domains for this system. Consider now systems where the spherical and dipolar species are of similar volume, so \( b / U \ll 1 \). For example, \( b / U \approx 1/20 \) in KBr::CN (see ref. \([24]\)), which is an OG for \( n \approx 0.5 \). For \( n \approx 0.5 \) we obtain \( \xi \approx 10 \) and \( N_d \approx (U / b)^{1/2} \approx 1000 \). However, in the dilute regime b), because of the small exponent in eq. (5), \( \xi \approx 3 \), and \( N_d \approx 30 \) only, a large reduction!

As mentioned above, the electric dipole interaction is typically quite small. For our purposes here, it renormalizes the Ising interaction, so that

\[
\tilde{U}_e \approx n \sqrt{\frac{\gamma^2}{\rho c^2} + U_e^2}
\]

and allows the coupling to an electric field \( E \). The latter allows a measurement of the effective RF as function of \( n \), providing an experimental check to our theory. For \( E > 0 \)

\[
\xi_E \approx \left( \frac{\bar{U}}{\sqrt{\bar{U}} + (E / d)^2} \right)^{(3/2)-\eta}
\]

where \( d \) is the dipole moment, \( b_{\text{eff}} = b_0 \sqrt{n} \) in regime a) and \( b_{\text{eff}} = \sqrt{\bar{U}} \) in regime b). Thus, increasing \( E \) causes a crossover from impurity-dominated to field-dominated \( \xi \) at \( E \cdot d \approx b_{\text{eff}} \). In fig. 1 we plot \( \xi \) for KBr::CN, for \( n = 0.5 \) and for \( n = 10^{-3} \approx b / U \). For \( E \gg b_{\text{eff}} / d \), one has \( \xi \approx E^{-1} (3/2-\eta) \). However, the magnitude of \( \xi_E \) at \( E = 0 \), as well as the region where its functional form deviates considerably from the above power law, are strongly \( n \)-dependent. Their measurement as functions of \( n \) would measure the effective RF in the system as function of dilution, and thus provide a check to our theory.

Extrinsic impurities also generate RFs with a broad distribution, and can be analyzed along the same lines as the intrinsic impurities. Thus, at \( E = 0 \), as the system is purified, the magnitude of \( \xi \) and of the cusp in the nonlinear permittivity increase, but only to a value in accordance with eqs. (4), (5).

**Spin glasses.** – We have seen that the random fields generated in OGS and DGs prevent a genuine phase transition to a low-\( T \) glassy state. Why does one then see a glass transition in SGSs? After all, one could certainly expect similar random fields to be generated therein, via, e.g., magnetoacoustic interactions. However, there is an essential difference between electric and magnetic Ising systems. If one neglects the volume term in eq. (2), then OGS share with SGs a symmetry under \( \sigma_i \rightarrow -\sigma_i \). However this is a symmetry under parity \( \hat{P} \) in OGS (where the
pseudospin variables $\sigma_z$ are not real spins), but under time-reversal $T$ in SGs (where the $\sigma_z$ are real spins, and couple to real magnetic fields). As a result, terms linear in $\sigma$ which emerge naturally in OGs, are not allowed in the absence of a magnetic field in SGs\textsuperscript{1}. For example, in zero field the magnetoacoustic interaction is, to lowest order, bilinear in $\sigma$:

$$V_{sp} = -\sum_j \sum_{\alpha\beta} \left[ \eta^{\alpha\beta} + \sum_\gamma \gamma^\alpha \gamma^\beta \right] \frac{\partial X_{j,\alpha}}{\partial x_{j,\beta}}, \quad (8)$$

where $A^{\alpha\beta\gamma\delta}_j$ is the spin-phonon interaction tensor. Thus, SGs are well described by the Edwards-Anderson model but by itself keeps the $H_1$, and thus, RFs will always emerge (this could be done, e.g., in the same way as suggested above for OGs, by mapping $b \leftrightarrow h$ and $E \leftrightarrow H_1$, where $H_1$ is the longitudinal magnetic field).

All our considerations for the emergence of effective RFs, both in OGs and in magnetic systems with transverse magnetic field, are independent of the thermodynamic phase of the system, i.e. they also apply to the ferromagnetic/ferroelectric phases [19]. Thus, we argue that in disordered system with no time-reversal symmetry the Ising model is unstable to small perturbations, and is therefore not realizable (i.e., RFs will always emerge). In particular, one cannot realize the transverse field Ising model in easy-axis disordered magnetic systems by applying a transverse magnetic field.

In light of these conclusions for SGs, it is interesting to recall that models of 2-level systems (TLSs) interacting with phonons are commonly used to describe a wide variety of glasses at low $T$, not just OGs and DGs. Insofar as the models of TLSs are applicable, and the volume term in the interaction with phonons causes a breaking of the $\sigma_z \leftrightarrow -\sigma_z$ symmetry, then our theory predicts that in these systems RFs will emerge as well, and there will also be a crossover rather than a phase transition between the glass-ordered and disordered phases. To unambiguously test this, one could measure the nonlinear dielectric susceptibility in these systems —it should not diverge as a function of temperature. These considerations apply both for the glass transition, and for the low-energy regime of interacting TLSs\textsuperscript{2}. For a recent calculation of the strength of the TLS-TLS interactions in the two regimes in orientational glasses, and a discussion of its applicability in structural glasses, see ref. [31].

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\textsuperscript{2}A transition to coherent orbital motion involving pairs of TLS coupled to nuclear quadrupolar moments is apparently seen at mK in some dipolar glasses (see refs. [29,30]). However this is not a glass transition.
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