Atomistic simulations of magnetic amorphous solids: Magnetostriction, Barkhausen noise and novel singularities

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Abstract – We present results of atomistic simulations of a new model of a magnetic amorphous solid subjected to mechanical strains and magnetic fields. Contrary to standard magnetic random systems which are studied on a lattice with random interaction, in the present approach all the randomness comes from the glassy nature of the material. The model employed offers new perspectives on important effects like plasticity and magnetostriction. It is shown that the plastic response in such systems exhibit singularities characterized by new exponents; the spatial structure of these plastic events requires a new coarse grained elasto-magnetic theory which is provided here.

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Our understanding of the mechanical properties of amorphous solids has received a strong boost from atomistic simulations, leading to novel theories of plastic behavior in such systems. In particular localized plastic events could be understood as the appearance of effective Eshelby inclusions in the elastic matrix [1,2]. This relation allows analytic derivations, culminating recently with a theory of shear localization in amorphous solids, leading to shear bands and material failure [3,4]. As long as only mechanical properties are involved, the nature of the plastic singularities is now understood, being dominated by a sequence of simple saddle node bifurcations as the material is strained. This leads to a high degree of universality in the nature of plastic events in widely different amorphous solids ranging from Lennard-Jones binary mixtures to metallic glasses [5].

In contrast, atomistic simulations of strained magnetic amorphous solids in the presence of magnetic fields are at an earlier stage of development. Theoretical considerations indicate that new singularities and richer physics are expected from the presence of two independent control parameters, mechanical and magnetic [6]. In this letter we report on such simulations that indeed appear to present much new physics. Here we discuss results on athermal quasi-static (AQS) simulations in two dimensions. Extensions to 3D and to finite temperature and strain rates will be reported elsewhere.

The model. – The model we employ is in the spirit of the Harris, Plischke and Zuckerman (HPZ) Hamiltonian [7] but with a number of important modifications to conform with the physics of amorphous magnetic solids [6]. First, Our particles are not pinned to a lattice. We write the Hamiltonian as

$$U(\{r_i\}, \{S_i\}) = U_{\text{mech}}(\{r_i\}) + U_{\text{mag}}(\{r_i\}, \{S_i\}),$$

(1)

where \(\{r_i\}_{i=1}^N\) are the 2-D positions of \(N\) particles in an area \(L^2\) and \(S_i\) are spin variables. The mechanical part \(U_{\text{mech}}\) is chosen to represent a glassy material with a binary mixture of 65% particles A and 35% particles B, with Kob-Anderson Lennard-Jones potentials [8] having a minimum at positions \(\sigma_{AA} = 1.17557, \sigma_{AB} = 1.0\) and \(\sigma_{BB} = 0.618034\) for the corresponding interacting particles. These values are chosen to guarantee good glass formation and avoidance of crystallization. The energy parameters chosen are \(\epsilon_{AA} = \epsilon_{BB} = 0.5 \epsilon_{AB} = 1.0\) in units for which the Boltzmann constant equals unity. All the potentials are truncated at distance 2.5\(\sigma\) with two continuous derivatives. Particles A carry spins \(S_i\); the B particles are not magnetic. The total particle density in our fixed volume was chosen to be \(\rho = 0.972\).

The magnetic potential needs to be modeled to best fit a particular material, and different materials will have somewhat different magnetic interactions. For concreteness we consider here the spins \(S_i\) to be classical \(xy\) spins; the orientation of each spin is then given by an angle \(\phi_i\). We also denote by \(\theta_i(\pi)\) the local preferred easy axis of anisotropy, and end up with the magnetic contribution to
the potential energy in the form [6]

\[
U_{\text{mag}}(\{r_i\}, \{S_i\}) = -\sum_{<ij>} J(r_{ij}) \cos (\phi_i - \phi_j) - \sum_i K_i \cos (\phi_i - \theta_i(\{r_i\}))^2 - \mu_A B \sum_i \cos (\phi_i). \tag{2}
\]

Here \( r_{ij} \equiv |r_i - r_j | \) and the sums are only over the \( A \) particles. For a discussion of the physical significance of each term the reader is referred to ref. [6]. It is important however to stress that in our model (in contradistinction with the HPZ Hamiltonian [7] and also with the Random Field Ising Model [9]), the exchange parameter \( J(r_{ij}) \) is a function of a changing inter-particle position (either due to external strain or due to non-affine particle displacements, and see below). We choose for concreteness the monotonically decreasing form \( J(x) = J_0 f(x) \) where \( f(x) \equiv \exp(-x^2/0.28) + H_0 + H_2 x^2 + H_4 x^4 \) with $\mu_0 = -5.51 \times 10^{-8}$, $H_2 = 1.68 \times 10^{-8}$, $H_4 = -1.29 \times 10^{-9}$. This choice cuts off \( J(x) \) at \( x = 2.5 \) with two smooth derivatives. In our case \( J_0 = 1 \).

Another important difference is that in our case the local axis of anisotropy \( \theta_i \) is not random, but is determined by the local structure: define the matrix \( T_i \) as

\[
T_i^{\alpha \beta} \equiv \sum_j J(r_{ij}) r_{ij}^{\alpha}r_{ij}^{\beta} / \sum_j J(r_{ij}). \tag{3}
\]

The matrix \( T_i \) has two eigenvalues in two dimensions that we denote as \( \kappa_{i,1} \) and \( \kappa_{i,2} \). The eigenvector that belongs to the larger eigenvalue \( \kappa_{i,1} \) is denoted by \( \vec{\eta}_i \). The easy axis of anisotropy is given by \( \theta_i \equiv \sin^{-1}(|\vec{\eta}_i|) \). Finally the coefficient \( K_i \) which now changes from particle to particle is defined as

\[
K_i \equiv \tilde{C} \left[ \sum_j J(r_{ij}) \right]^2 (\kappa_{i,1} - \kappa_{i,2})^2, \quad \tilde{C} = 5.0/J_0 \sigma_{AB}^4. \tag{4}
\]

This definition, though not unique, guarantees both that \( K_i \) has units of energy and that the contribution due to anisotropy will vanish when the local neighborhood of the \( i \)-th particles is isotropic. The last term in eq. (2) is the interaction with the external field \( B \). We have chosen \( \mu_A \) in the range \([-0.08, 0.08]\). At the two extreme values all the spins are aligned along the direction of \( B \).

**New exponents.** To initiate the study of the interesting physics exhibited by this model we show in fig. 1 how the energy changes when the magnetic field increases without external strain. The increase of \( B \) is done quasi-statically, performing energy minimization after every step of increase of \( B \). Reversible smooth changes in the energy are punctuated by (irreversible) drops in energy which are caused by localized magneto-plastic events. As usual, these events are associated with an eigenvalue of the Hessian matrix going to zero, while at the same time the associated eigenfunction that is delocalized far from the instability gets localized on \( n \) particles where \( n \) can be much smaller and independent of \( N \) or it may scale like \( N^a \) depending on the value of \( B \) [10,11]. As stated above, in amorphous solids subjected to pure mechanical strains the lowest eigenvalue \( \lambda_P \) goes to zero with a characteristic \( \nu = 1/2 \) exponent [12], i.e. \( \lambda_P \sim (\gamma_p - \gamma)^{1/2} \) where \( \gamma \) is the magnitude of the external strain and \( \gamma_p \) its value at the occurrence of the plastic event [5]. Our first interesting finding is that this is no longer the case here, and \( \nu \) can differ from 1/2. In fig. 2 we show log-log plots of \( \lambda_P \) vs. \( B_p - B \) close to an instability at \( B_P \), indicating a complex critical behavior \( \lambda_P \sim (B_p - B)^\nu \) with exponents \( \nu \) that can differ from 1/2. For \( B_P \) very small and very large (see, for example, panel (a)) the slope in the log-log plot is very close to 1/2. There is a value of \( B_P \), which we denote as \( B_c \), for which we expect a slope of 3/4. In panel (b), \( B_p \) is in the vicinity of \( B_c \) and \( \nu \approx 0.71 \). For \( 0 < B_p < B_c \) and \( B_p > B_c \) we expect a crossover between a slope higher than 1/2 to a slope 1/2 in the vicinity of \( B_p \).

To understand these results we recall that the eigenvalue that goes soft belongs to the Hessian matrix \( \mathbf{H} \) which in the present case takes on the form [6]:

\[
\mathbf{H} = \begin{pmatrix}
\frac{\partial^2 U}{\partial \phi_i \partial \phi_j} & \frac{\partial^2 U}{\partial \phi_i \partial \phi_j} \\
\frac{\partial^2 U}{\partial \phi_i \partial \phi_j} & \frac{\partial^2 U}{\partial \phi_i \partial \phi_j}
\end{pmatrix}.
\tag{5}
\]

The eigenfunctions associated with the eigenvalues \( \lambda_k \) are denoted \( \Psi^k \). In terms of these objects we have an exact result for the change of \( \lambda \) with \( B \) [6]:

\[
\frac{\partial \lambda_k}{\partial B} |_{\gamma} = c_k^{(b)} \sum_i \frac{a_k^{(b)} [a_k^{(c)} b_{kkf} + b_k^{(c)}]}{\lambda_k}.
\tag{6}
\]

The precise definition of all the coefficients is given explicitly in ref. [6]. For the present purpose it is sufficient to know that \( c_k^{(b)} \) and both \( b_{kkf} \) cannot be zero or singular,
There exists a value of $B$ (panel (a) is for large $B$ for different values of $B$ but on the other hand $a$ for the lowest eigenvalue of the Hessian matrix Fig. 2: (Colour on-line) Scaling laws and apparent scaling laws instability (panel (c)).

$$\ell \equiv \frac{b}{\Xi(b)\Psi(b)} \equiv \frac{\partial^2 U}{\partial B \partial \phi}. \quad (7)$$

but on the other hand $a^{(b)}$ may vanish at some value of $B$. This coefficient is defined as

Clearly, when one of the eigenvalues $\lambda_p$ goes to zero, eq. (6) simplifies to one dominant term

$$\frac{\partial \lambda_p}{\partial B} \bigg|_0 \approx -\frac{a_p^{(b)}C}{\lambda_p}. \quad (8)$$

Where $C$ is a constant when $B \to B'_p$. We have checked that in our system there exists a gap to the next eigenvalue justifying the last equation. Examining our Hamiltonian we discover that

$$a_p^{(b)} = \sum_{i=1}^{N} \sin \phi_i \Psi_i^p. \quad (9)$$

Noting that $\Psi^{(p)}$ gets localized on $n \ll N$ particles and that it is normalized, we expect

$$a_p^{(b)} \approx \frac{1}{\sqrt{n}} \sum_{i=1}^{n} \sin \phi_i = \sqrt{n} (\sin \phi)_n, \quad (10)$$

where the notation $\langle \ldots \rangle_n$ means an average over the particles on which the eigenfunction is localized. From this result we conclude that whenever $(\sin \phi)_n$ is not zero, eq. (8) will lead to exponent $\nu = 1/2$. Indeed for small $B$ the spins point out in the quasi-random local anisotropy axis, and $(\sin \phi)_n \neq 0$. Also when $B$ is very large, most of the spins point out in the $x$ direction, but unstable modes will consist of spins pointing otherwise, so that again $(\sin \phi)_n \neq 0$. But for an intermediate value of $B$, where the drops in $U$ in fig. 1 or in the magnetization in fig. 4 are largest, the most unstable mode will consist of spins pointing almost opposite to the applied field, with $\phi_i \approx \pi$. We thus define the value of $B = B_c$ as that point for which (in the thermodynamic limit) $\langle \sin \phi \rangle_n = 0$. For $B_p$ in the vicinity of $B_c$ we write $\phi_i = \pi + \Delta \phi_i$, and therefore

$$a_p^{(b)} \approx -\frac{1}{\sqrt{n}} \sum_{i=1}^{n} \sin \Delta \phi_i \approx -\frac{1}{\sqrt{n}} \sum_{i=1}^{n} \Delta \phi_i \approx \sqrt{\langle (\Delta \phi_i)^2 \rangle_n}, \quad (11)$$

by the central limit theorem. Since $\langle (\Delta \phi_i)^2 \rangle_n = 0$ at $B = B_c$ we expect in the vicinity of $B_c$ to have $\langle (\Delta \phi_i)^2 \rangle_n \propto (B_c - B)$. Substituting this in eq. (8) we get immediately $\nu = 3/4$ as is seen in the numerics. Note that this result is only relevant for $B_c$. At any other value of $B$ we expect to see $\nu = 1/2$ or a crossover to this value for $B \to B'_p$.

Magnetostriiction. – Another interesting physical effect that warrants much more analysis is magnetostriiction. This effect can be studied in an NPT ensemble as the change in the volume with changing the magnetic field or in an NVT ensemble by the change in the pressure. We chose here the latter and in fig. 3 we display the pressure $P$ as a function of $B$ in a quasi-static ramp of $B$. During smooth sections the magnetostriiction coefficient is positive in our case (in the NVT ensemble the pressure increases with $B$) but the pressure is again punctuated by plastic drops which lead to an overall decrease in pressure.
coordinate change, proportional to \( H \) is singular however at a plastic event, being a non-affine plastic drop with a slope \( 1/2 \).

Between the drops the pressure increases with a tendency towards singularity immediately preceding the drop. To understand the physics displayed here we recall that using the virial theorem the pressure is written in our system (at \( T = 0 \)) as

\[
P = \frac{1}{2V} \sum_{i\neq j} f_{ij} \cdot r_{ij}, \quad r_{ij} = r_i - r_j, \tag{12}
\]

where \( f_{ij} \) is the force exerted by the \( j \)-th particle on the \( i \)-th particle. Taking the derivative with respect to \( B \),

\[
2V \frac{\partial P}{\partial B} = \sum_{i\neq j} \frac{\partial f_{ij}}{\partial B} \cdot r_{ij} + \sum_{i} f_{ij} \frac{\partial r_{ij}}{\partial B}. \tag{13}
\]

The first term on the RHS is not expected to be singular when \( B \to B_p \), since the derivative there is of the type of \( \Xi \) of eq. (7) which is never singular. The second term is singular however at a plastic event, being a non-affine coordinate change, proportional to \( H^{-1} \cdot \Xi \). Near the plastic event this second term is proportional to the RHS of eq. (8), so that for \( B \) small or large we expect

\[
P \approx P_p - C_1 (B_p - B) - C_2 \sqrt{B_p - B}, \tag{14}
\]

where \( P_p \) is the value of the pressure before the plastic drop, and \( C_1 \) and \( C_2 \) positive. A fit to this formula for \( B_p \approx 0.0044 \) is shown in the insets in fig. 3, explaining the shape of the smooth parts and the plastic punctuations in fig. 3. For \( B_p \) close to \( B \), the new exponent \( \nu = 3/4 \) will change eq. (14) in an obvious way.

**Barkhausen noise.** – Needless to say, our model allows for a detailed study of the Barkhausen noise [13]. In fig. 4 we exhibit the magnetization as a function of \( B \). The shorter line represents the change in magnetization as \( B \) is increased starting from the freshly quenched glass. Upon saturation, the magnetic field is inverted until the magnetization becomes \( -1 \), where the field is again increased to display the hysteresis loop. The statistics of Barkhausen noise was studied extensively in the context of the Random Field Ising Model with strong claims for universality [14]. The model proposed here promises excellent testing grounds of such claims in a model of actual amorphous solids with magnetic properties. It goes beyond the scope of this letter to present the statistics of the Barkhausen noise, and we will do it in a forthcoming paper.

**Coarse-grained model.** – Finally we should comment on the interesting subject of the non-affine displacements in this model and how to provide a theory for them. In amorphous solids undergoing purely mechanical loading the plastic events are well described by the Eshelby theory, being quadrupolar in 2D with obvious generalization to 3D. In the present case the mechanical part of the non-affine displacements are still quadrupolar, but the magnetic non-affine changes are totally different, and may even display magnetic topological singularities. An example of a typical event that displays \( \Delta S_i \) in a plastic event is shown in fig. 5. To create the analog of the Eshelby theory for those we need first to generalize the Lamé-Navier equations to the present case. A convenient starting point is the free energy functional

\[
F = \int \text{d}x \text{d}y \left[ \mu (u_{ik} - \frac{1}{2} \delta_{ik} u_{ff})^2 + \frac{1}{2} \kappa u_{ff}^2 - K (m_i m_i)^2 - b \left( u_{ik} - \frac{u_{ff}}{2} \delta_{ik} \right) m_i m_k + \frac{a}{2} \left( \frac{\partial m_i}{\partial x_k} \right)^2 - B m_z \right], \tag{15}
\]
where $\mu$ and $\kappa$ are the shear and bulk moduli and $b$ is the anisotropic magnetoelastic coupling term. $\mathbf{n}$ is a random vector field representing the local anisotropy. The coupling constant $a$ represents the exchange interaction. For an $xy$ model $m_x = \cos \theta$ and $m_y = \sin \theta$. Substituting and following the Euler-Lagrange procedure one ends up with the generalized Lamé-Navier equations

$$
\mu \left( \frac{\partial^2 u_x}{\partial x^2} + \frac{\partial^2 u_x}{\partial y^2} \right) + \kappa \left( \frac{\partial}{\partial x} \left( \frac{\partial u_x}{\partial x} + \frac{\partial u_y}{\partial y} \right) \right) = \\
k \left[ \cos 2\theta \left( \frac{\partial \theta}{\partial y} \right) - \sin 2\theta \left( \frac{\partial \theta}{\partial x} \right) \right],
$$

$$
\mu \left( \frac{\partial^2 u_y}{\partial x^2} + \frac{\partial^2 u_y}{\partial y^2} \right) + \kappa \left( \frac{\partial}{\partial x} \left( \frac{\partial u_x}{\partial x} + \frac{\partial u_y}{\partial y} \right) \right) = \\
k \left[ \cos 2\theta \left( \frac{\partial \theta}{\partial x} \right) + \sin 2\theta \left( \frac{\partial \theta}{\partial y} \right) \right],
$$

(16)

$$
a \left[ \frac{\partial^2 \theta}{\partial x^2} + \frac{\partial^2 \theta}{\partial y^2} \right] = k \left[ \frac{\partial u_x}{\partial x} - \frac{\partial u_y}{\partial y} \right] \sin 2\theta
$$

$$
- \left( \frac{\partial u_x}{\partial y} + \frac{\partial u_y}{\partial x} \right) \cos 2\theta + B \sin \theta
$$

$$
+ K \sin 2(\theta - \eta(x, y)),
$$

with $\eta(x, y)$ being the angle of the local easy axis that varies randomly. The first two equations simplify to the standard Lamé-Navier equations when $k = 0$. The geometric characters of the non-affine plastic events are determined by these equations.

In summary, we have offered a new model for an amorphous solid with magnetic interactions. Contrary to previous work concerning magnetic random interactions which were studied on a simple lattice but with random interactions, we constructed above a model in which the randomness appears due to the glassy nature of the material. The results of this novel approach indicate new physics. Firstly, the exponents characterizing the plastic instabilities change from the respective regular atomic glass. In that case the exponent $\nu = 1/2$ was determined by the simple saddle node bifurcation associated with the plastic event. The richer plastic event in the present case allows changes in these exponents, up to $\nu = 3/4$. Second, we find that magnetostriction is also associated with non-trivial changes of the pressure upon changing the magnetic field, with square-root singularity approach towards the sharp plastic events. This prediction can be already put to experimental tests. Finally, the model exhibits Barkhausen noise, but the statistics of the latter will be fully discussed in a future publication. It is our aim to elucidate all these new aspects in full detail in forthcoming publications.

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REFERENCES

[1] MALANDRO D. L. and LACKS D. J., J. Chem. Phys., 110 (1999) 4503.
[2] MALONEY C. and LEMAIître A., Phys. Rev. Lett., 93 (2004) 195501; 016001; J. Stat. Phys., 123 (2006) 415.
[3] DASGUPTA R., HENTSCHEL H. G. E. and PROCACCIA I., Phys. Rev. Lett., 109 (2012) 25502; Phys. Rev. E, 87 (2013) 022810.
[4] ASHWIN J., GENDelman O., PROCACCIA I. and SHOR C., Atomistic theory of the shear band direction in amorphous solids, submitted to Phys. Rev. Lett., arXiv:1304.4009.
[5] DASGUPTA R., KARMAKAR S. and PROCACCIA I., Phys. Rev. Lett., 108 (2012) 075701.
[6] HENTSCHEL H. G. E., ILYIN V. and PROCACCIA I., EPL, 99 (2012) 26003.
[7] HARRIS R., Plishcke M. and ZUCKERMAN M. J., Phys. Rev. Lett., 31 (1973) 160.
[8] KOB W. and ANDERSEN H. C., Phys. Rev. E, 51 (1995) 4626.
[9] SETINA J. P., DAHMEK K., KATHA S., KRUHMANSL J. A., ROBERTS B. W. and SHORE J. D., Phys. Rev. Lett., 70 (1993) 3347.
[10] LERNER E. and PROCACCIA I., Phys. Rev. E, 79 (2009) 066109.
[11] PERKOVIC O., DAHMEK K. A. and SETINA J. P., Phys. Rev. B, 59 (1999) 6106.
[12] KARMAKAR S., LERNER E. and PROCACCIA I., Phys. Rev. E, 82 (2010) 051103(R).
[13] LE DOUSSAL P., MÜLLER M. and WIESE K. J., EPL, 91 (2010) 57004.
[14] PERKOVIC O., DAHMEK K. A. and SETINA J. P., Phys. Rev. Lett., 75 (1995) 4528.

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