Magnetomechanical coupling in thermal amorphous solids

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Standard approaches to magnetomechanical interactions in thermal magnetic crystalline solids involve Landau functionals in which the lattice anisotropy and the resulting magnetization easy axes are taken explicitly into account. In glassy systems one needs to develop a theory in which the amorphous structure precludes the existence of an easy axis, and in which the constituent particles are free to respond to their local amorphous surroundings and the resulting forces. We present a theory of all the mixed responses of an amorphous solid to mechanical strains and magnetic fields. Atomistic models are proposed in which we test the predictions of magnetostriction for both bulk and nanofilm amorphous samples in the paramagnetic phase. The application to nanofilms with emergent self-affine free interfaces requires a careful definition of the film "width" and its change due to the magnetostriction effect.

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I. INTRODUCTION

The subject of the interaction between mechanical and magnetic properties in magnetic glasses has been relatively neglected by theorists. Despite the enormous amount of work on magnetism in crystalline materials (including "spin glasses" where spins are restricted to reside on a lattice), and the equally enormous amount of work on nonmagnetic glasses, there have been almost no theoretical studies of elastic, plastic, and magnetic responses to shear and to external magnetic fields in glasses with magnetic properties until recently [1–7]. The crucial difference is that particles in a glass are free to move around whether they carry spins or not, and therefore there is a strong coupling between the mechanical and the magnetic properties of these materials. A generic plastic event in such materials is accompanied by simultaneous discontinuous changes in stress, energy, and magnetization, cf. Fig. 1. A number of model glasses with magnetic interaction were put forward, allowing highly accurate simulations for which one could offer detailed theories [4–7]. It was shown that magnetism can be induced by plastic events [6]. One could also study with exquisite detail the statistics of Barkhausen noise in magnetic glasses to discover that it can belong to a number of different universality classes depending on the details of the magnetic interactions [8].

One of the best known and important cross effects between mechanics and magnetism is magnetostriction (shape change due to to applied magnetic fields) and its inverse, the Villari effect (induced magnetization due to mechanical strain). The theory of magnetostriction as applied to solids and thin films typically makes a number of implicit assumptions about the solids investigated. Often one presumes that the considered solid has a crystalline structure and the magnetic atoms lie at well defined lattice sites (up to thermal fluctuations). In consequence, due to spin-orbit coupling, there exist global magnetic easy axes along which the macroscopic magnetization prefers to orient at low temperatures. In the absence of such easy axes, if the only magnetic interaction is an exchange interaction, all directions are degenerate in the absence of an applied magnetic field. Usually the assumption is also made that we are dealing with a low temperature situation in which the magnetization is saturated in magnitude and thus the easy axis controls the angle but not the magnitude of the magnetization **m**. If these were the only magnetic energy terms there would not exist any magnetostriction. But there also exists strain energy in the solid and this is coupled to the magnetization, resulting in a strained configuration of the solid as the lowest energy state of the system. This can be seen in the easiest way if we write down the Landau function *F* for the magnetic and strain energies. Taking for example a cubic lattice at low temperatures $T \ll T_c$ with saturated magnetization,

$$F = K (\alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2) + B_1 (\alpha_1^2 \epsilon_{xx} + \alpha_2^2 \epsilon_{yy} + \alpha_3^2 \epsilon_{zz}) + B_2 (\alpha_1 \alpha_2 \epsilon_{xy} + \alpha_2 \alpha_3 \epsilon_{yz} + \alpha_3 \alpha_1 \epsilon_{zx}) + (1/2)c_{11} (\epsilon_{xx}^2 + \epsilon_{yy}^2 + \epsilon_{zz}^2) + (1/2)c_{44} (\epsilon_{xy}^2 + \epsilon_{yz}^2 + \epsilon_{zx}^2) + c_{12} (\epsilon_{yy} \epsilon_{zz} + \epsilon_{xx} \epsilon_{zz} + \epsilon_{xx} \epsilon_{yy}).$$
(1)

Here α_i are the cosines of the angles between the magnetization direction and the cube easy axes x_i , while ϵ_{ij} is the strain tensor. *K* is the strength of the anisotropy energy, B_1 and B_2 are magnetoelastic coupling constants, while c_{ij} are the elastic moduli of the cube. By minimizing the Landau functional with respect to strain $\partial F / \partial \epsilon_{ij} = 0$ we can find explicit expressions for the strain in the material that minimizes the energy

$$\epsilon_{ii} = \frac{B_1 [c_{12} - \alpha_i^2 (c_{11} + 2c_{12})]}{[(c_{11} - c_{12})(c_{11} + 2c_{12})]}$$

$$\epsilon_{ij} = \frac{-B_2 \alpha_i \alpha_j}{c_{44}}.$$
(2)

Using Eq. (2), the magnetostriction in a direction $\beta \equiv (\beta_1, \beta_2, \beta_3)$ where the β_i are the cosines of the angles between the measurement direction and the cube axes x_i is then



FIG. 1. Energy, stress, and magnetization changes during plastic events as a result of increasing strain in a magnetic amorphous solids. The data is taken from Ref. [9] for an athermal example.

given by

$$\delta \ell / \ell = \sum_{i \leqslant j} \epsilon_{ij} \beta_i \beta_j.$$
(3)

This result was first derived by N. Akulov in 1926 [10].

In this paper we focus on situations involving amorphous solids and metallic glasses, where the basic assumptions made in deriving Eqs. (1) and (2) do not apply as in amorphous solids there is typically no global easy axis in the material. Moreover, we are often interested in situations where we are in a glass phase $T < T_g$ but above or close to the Curie temperature $T \approx T_c$ so that we cannot assume either that the magnetization is saturated or that applied magnetic fields **B** do not have a strong influence on the size of the magnetization **m**. Finally Eq. (1) is a macroscopic energy functional and we would like to look at magnetostriction in a more microscopic (atomistic) context in glasses.

The structure of this paper is as follows: In Sec. II we present atomistic models for magnetic glasses. In Sec. III we describe the general approach to the responses of magnetic glasses to mechanical and magnetic strains. In Sec. IV the numerical simulations are presented, stressing results for magnetostriction in both bulk and film glasses. Section V applies the general results of Sec. III to extracting the magnetostricition coefficient in thermal glassy materials. Section VI provides a summary of the paper and some discussion.

II. MICROSCOPIC MODELS

The potential energy U of N point particles in an amorphous magnetic solids in the presence of a magnetic field B can be written as

$$U(\{\mathbf{r}_i\},\{\mathbf{S}_i\}) = U_{\text{mech}}(\{\mathbf{r}_i\}) + U_{\text{mag}}(\{\mathbf{r}_i\},\{\mathbf{S}_i\};\mathbf{B}), \quad (4)$$

where $\{r_i\}_{i=1}^N$ are the positions of the particles and S_i are spin variables.

A. The mechanical interactions

The mechanical part of our Hamiltonian Eq. (4) can be taken as any of the standard models of glass formers, and we will assume that it is a sum of binary interactions such that

$$U_{\text{mech}}(\{\boldsymbol{r}_i\}) = \sum_{\langle ij \rangle} \phi(r_{ij}), \qquad (5)$$

where $\langle ij \rangle$ means "all distinct pairs," $r_{ij} \equiv |\mathbf{r}_i - \mathbf{r}_j|$ are the instantaneous distances between particles *i* and *j*. This still leaves a lot of freedom, as the literature attests to a variety of models with binary interactions that produce good glass formers. To generate a glass, we simulate a Kob-Andersen binary mixture of two types of particles [11–13]. We address the particles as type-A particles which are magnetic and the other type of particles, which are nonmagnetic, as type-B particles. The ratio of the number of *A* and *B* type particles is taken as 80 : 20. The mechanical part of interatomic interactions is defined by truncated and shifted Lennard-Jones potentials

$$\phi_{ij}(r) = \begin{cases} \phi_{ij}^{LJ}(r) + C_{ij} & \text{if } r \leq R_{ij}^{\text{cut}}, \\ 0 & \text{if } r > R_{ij}^{\text{cut}}, \end{cases}$$
(6)

where $C_{ij} = -\phi_{ij}^{LJ}(R_{ij}^{\text{cut}})$ and

$$\phi_{ij}^{LJ}(r) = 4e_{ij} \left[\left(\frac{\sigma_{ij}}{r} \right)^{12} - \left(\frac{\sigma_{ij}}{r} \right)^6 \right].$$
(7)

To simplify the simulations the pair interactions in Eq. (6) are truncated at distance $R_{ij}^{cut} = 2.5\sigma_{ij}$. It is convenient to introduce reduced units, with σ_{AA} being the unit of length and e_{AA} the unit of energy. Parameters for *A*-*B* and *B*-*B* interactions are given by $\sigma_{BB}/\sigma_{AA} = 0.88$, $\sigma_{AB}/\sigma_{AA} = 0.8$, $e_{BB}/e_{AA} = 0.5$, and $e_{AB}/e_{AA} = 1.5$. The reported glass transition temperature T_g of the Kob-Andersen binary mixture in 3D [13] is $T_g = 0.28$.

B. The magnetic interactions

The magnetic properties of amorphous magnets are extremely varied and cannot be represented by a unique Hamiltonian. For example spins can be effectively localized on individual atoms, or be dominated by delocalized spins on conduction electrons. If localized the total angular momentum of unpaired electrons will depend on the atomic species considered. In this paper we shall consider the simplest Heisenberg magnetic Hamiltonian that couples magnetic and mechanical properties in amorphous solids—namely an exchange interaction in which the exchange integral is an explicit function of particle positions

$$U_{\text{mag}}(\{\boldsymbol{r}_i\},\{\boldsymbol{S}_i\};\boldsymbol{B}) = -\sum_{\langle ij\rangle} J(r_{ij}) \mathbf{S}_i \cdot \mathbf{S}_j - g\mu_B \sum_i \mathbf{S}_i \cdot \mathbf{B}.$$
(8)

The first term on the right hand side is the short range exchange interaction $U_{\text{ex}}(\{r_i\},\{S_i\})$. Typically the exchange integral J(r) > 0, thus encouraging ferromagnetism, and will be peaked at some distance r_1 or else be an exponentially decreasing function of r. Note that $J(r_{ij})$ will couple magnetism and strain in a nontrivial fashion. The exchange energy

$$J(x) = J_0 \frac{\exp(-\kappa x)}{x}.$$
 (9)

Like the Lennard-Jones interaction, the exchange interaction is also truncated at x = 2.5 and shifted to zero at that point. The screening parameter κ determines the range of the interaction. We have taken $\kappa = 3.6$. Finally, in our case $J_0 = 3.0$.

III. MECHANICAL AND MAGNETIC RESPONSES AT FINITE TEMPERATURES

The theory of mechanical and magnetic responses of amorphous solids at zero temperature is available, and for completeness we summarize the main results in Appendix B. Here we present the theory for thermal glasses, taking into account the effects of thermal fluctuations. Given any dynamical variable $Y(\{r_i, S_i\}_{i=1}^N)$ its thermal average is determined by

$$\langle Y \rangle = \frac{\int dX dSY e^{-\beta U(X,S)}}{Z}$$
$$Z = \int dX dS e^{-\beta U(X,S)}$$
$$X \equiv \{\mathbf{r}_i\}_{i=1}^N, \quad S \equiv \{\mathbf{S}_i\}_{i=1}^N.$$
(10)

Noticing that $\langle Y \rangle$ is a function of the magnetic field and the mechanical strain we can compute

$$\frac{\partial \langle Y \rangle}{\partial B_{\alpha}} = \left\langle \frac{\partial Y}{\partial B_{\alpha}} \right\rangle - \beta \left[\left\langle Y \frac{\partial U}{\partial B_{\alpha}} \right\rangle - \left\langle \frac{\partial U}{\partial B_{\alpha}} \right\rangle \langle Y \rangle \right],$$
$$\frac{\partial \langle Y \rangle}{\partial \epsilon_{\alpha\beta}} = \left\langle \frac{\partial Y}{\partial \epsilon_{\alpha\beta}} \right\rangle - \beta \left[\left\langle Y \frac{\partial U}{\partial \epsilon_{\alpha\beta}} \right\rangle - \left\langle \frac{\partial U}{\partial \epsilon_{\alpha\beta}} \right\rangle \langle Y \rangle \right]. \quad (11)$$

Specializing these equations to the stress tensor and the magnetization we get the explicit expressions for the magnetostriction and the Villari effects in thermal systems:

$$\frac{\partial \langle \sigma_{\alpha\beta} \rangle}{\partial B_{\gamma}} = \left\langle \frac{\partial \sigma_{\alpha\beta}}{\partial B_{\gamma}} \right\rangle - \beta \left[\left\langle \sigma_{\alpha\beta} \frac{\partial U}{\partial B_{\gamma}} \right\rangle - \left\langle \frac{\partial U}{\partial B_{\gamma}} \right\rangle \langle \sigma_{\alpha\beta} \rangle \right], \quad (12)$$

$$\frac{\partial \langle m_{\gamma} \rangle}{\partial \epsilon_{\alpha\beta}} = \left\langle \frac{\partial m_{\gamma}}{\partial \epsilon_{\alpha\beta}} \right\rangle - \beta \left[\left\langle m_{\gamma} \frac{\partial U}{\partial \epsilon_{\alpha\beta}} \right\rangle - \left\langle \frac{\partial U}{\partial \epsilon_{\alpha\beta}} \right\rangle \langle m_{\gamma} \rangle \right]. \quad (13)$$

Magnetostriction is actually determined by the strain response to the magnetization, but using linear elasticity theory we can easily invert from the stress response to the strain response, as

$$\sigma_{\alpha\beta} = c_{\alpha\beta\gamma\delta}\epsilon_{\gamma\delta}, \qquad (14)$$

where c is the elastic modulus tensor. Equations (12) and (13) can be simplified further by using the identities

$$\frac{\partial U}{\partial B_{\gamma}} = -Vnm_{\gamma}, \quad m_{\gamma} = \frac{g\mu_B}{N_s} \sum S_i$$
(15)

$$\frac{\partial U}{\partial \epsilon_{\alpha\beta}} = V \sigma_{\alpha\beta}.$$
 (16)

Here *n* is the number of spins per unit volume and N_s the number of particles carrying spins in the system. The reader should note that here m_{γ} and $\sigma_{\alpha\beta}$ are the instantaneous variable rather than thermal averages. Using these identities in Eqs. (12)

and (13) we get the final results

$$\frac{\partial \langle \sigma_{\alpha\beta} \rangle}{\partial B_{\gamma}} = \left\langle \frac{\partial \sigma_{\alpha\beta}}{\partial B_{\gamma}} \right\rangle + \beta V n [\langle \sigma_{\alpha\beta} m_{\gamma} \rangle - \langle m_{\gamma} \rangle \langle \sigma_{\alpha\beta} \rangle], \quad (17)$$

$$\frac{\partial \langle m_{\gamma} \rangle}{\partial \epsilon_{\alpha\beta}} = \left\langle \frac{\partial m_{\gamma}}{\partial \epsilon_{\alpha\beta}} \right\rangle - \beta V [\langle m_{\gamma} \sigma_{\alpha\beta} \rangle - \langle \sigma_{\alpha\beta} \rangle \langle m_{\gamma} \rangle].$$
(18)

While the first term in the r.h.s of Eqs. (17), also called the Born term [15], defines the affine part, the second term comes from the nonaffine fluctuation at finite temperature. The fluctuation term does not necessarily vanish for $T \rightarrow 0$. This is due to the fact that the particle displacements need not follow an imposed macroscopic strain affinely. While the fluctuation term gives the extent of these nonaffine displacements [16], the Born term reflects the affine part of the particle displacements.

From the identity of the mixed second derivatives of U we can derive immediately the Maxwell relation

$$\frac{\partial \langle \sigma_{\alpha\beta} \rangle}{\partial B_{\gamma}} = -n \frac{\partial \langle m_{\gamma} \rangle}{\partial \epsilon_{\alpha\beta}}.$$
(19)

In the next subsection we motivate further discussion by specializing to Lennard-Jones glass formers.

IV. NUMERICAL SIMULATIONS

The numerical creation of glassy bulk and film phases and their equilibration using Monte Carlo techniques is presented in Appendix A. We discuss separately the results for bulk and film.

A. Bulk phase

In the bulk phase, we simulate N = 4000 particles contained in a cell endowed with periodic boundary condition in all three directions to mimic an infinite system. The pressure and the temperature were fixed at P = 2.2 and T = 0.23 (below T_g but above T_c).

Taking the external field to point in the z direction, we define

$$s \equiv \left\langle \frac{\sum_{i} S_{i}^{z}}{N_{s}} \right\rangle. \tag{20}$$

This quantity was computed in an NPT ensemble and its dependence on the external field is shown in Fig. 2. At zero field the system is disordered and at high values of the field the magnetization saturates. In Monte Carlo simulations at fixed pressure the average volume changes with increasing the external field. This is measured by the changing length L of the simulation cell; the probability distribution function (pdf) P(L) at different values of applied external field h is shown in Fig. 3. In general, this magnetostriction effect is weak but clearly observable. The magnetostriction is quantified as the fractional change in length of the sample $\gamma = \delta L/L$. When γ is measured at high values of the external magnetic field (i.e., when the magnetization saturates) one refers to the "saturation magnetostriction." In fact it is advantageous to define the magnetostriction effect by its dependence on s (see, e.g., Ref. [17]). Indeed, the experimental results in Ref. [17] exhibit a linear dependence of the magnetostriction when plotted as a function of s^2 . The same result was obtained analytically using the model discussed in Ref. [18]. Results of



FIG. 2. Dependence of *s* on the applied magnetic field for the bulk magnetic glass. Here $h \equiv g\mu_B B$.

our simulations for the bulk phase are in agreement, cf. Fig. 4 where the dependence of L on s^2 is observed. Fitting the data by least squares and denoting the length of the simulation cell at zero magnetization L_0 the quantity $\gamma = (L_0 - L)/L_0$ was calculated. Therefore, one can write

$$\gamma = \lambda s^2. \tag{21}$$

B. Film construction

A nanothin film is generated on top of a face-centered cubic crystalline substrate composed of fixed $N_S = 1152$ identical particles interacting via Lennard-Jones potential with the film particles. The interaction parameters are: $\sigma_{SA}/\sigma_{AA} = \sigma_{SB}/\sigma_{AA} = 0.8$, $e_{SA}/\epsilon_{AA} = e_{SB}/e_{AA} = 1.5$. The subscript *S* stands for substrate.

The substrate particles are taken to be nonmagnetic. The substrate density in equilibrium is $\rho_S = 2.1$, providing a support to the film whose lateral dimensions are $L \times L$. To create a film of the binary mixture we return to our bulk simulation and cut off a slab of desired width w perpendicular to the *z* direction with lateral dimensions $L \times L$. The geometric fit of the film slab to the substrate is obtained by taking the bulk at density $\rho = 1.22$.

When the slab is positioned on the substrate it is placed with a gap between the film and the substrate which is $2^{1/6}\sigma_{SA}$



FIG. 3. P(L) in NPT simulations of the amorphous glass at different values of the applied magnetic field.



FIG. 4. Dependence of average value of L in NPT simulations s^2 .

at the minimum of the Lennard-Jones potential between the substrate and the *A* particles. The created simulation box is kept periodic in the *x* and *y* directions with the length of the periodicity cell begin L = 14.86. The substrate acts as a fixed wall at the bottom of the film. In order to create an equilibrated film we first impose a maximal extent of the film on the upper boundary. Switching on the Monte Carlo algorithm, when a particle attempts to cross the upper boundary, this move is rejected. The initial gap between the upper particles in the film and and this boundary is $2.5\sigma_{AA}$. Clearly, when the NVT Monte Carlo steps accumulate, the film adjusts the height along *z* creating an upper free boundary.

C. Computing the film width

Due to the existence of the substrate at the bottom (z = 0 is defined at the top of the crystalline substrate) and a free film at the top, translational invariance in the z direction is lost. Indeed, the local density along z for two films of different widths is shown in Fig. 5. Like in liquid phases (see Ref. [19]) the films show ordering near the substrate. In the wider film the region farther from the substrate reaches the average density in



FIG. 5. The local density as a function of z for two films containing different number of particles, N = 867 and N = 1940, respectively. The red constant line represents the average density of the bulk phase from which the film was created.



FIG. 6. Example of distributions of the local width for different size of subdomains $l \times l$ in a film at temperature T = 0.23.

the bulk phase. In both cases near the free boundary there is a smooth crossover from the dense phase to vacuum. This makes the influence of film topography on the film width nontrivial. For large films the surface will be self-affine. Here we are specifically interested in how to treat nanofilms in which the fluctuations of the free surface are of the order of the film width.

In general the width of a given film will depend on the number of particles N and the magnetization s. In order to define the film width, the area of the simulation cell is divided into subdomains of size $l \times l$. In each subdomain, one creates a list of particles ordered according to their z coordinate from top to bottom. After every Monte-Carlo sweep we determine again this ordering. In each realization the particle with the highest value of z in each subdomain determines the position of the free surface, defined as w_{loc} . The typical distributions of the local widths in a film of binary mixture for different l is shown in Fig. 6. The position of the maximum of this distribution defines "the width" of the film w(l,s). As one can see the estimated width of a film increases with increasing l. The dependence of the width on s measured at different values of l is shown in Fig. 7. Examining Fig. 7 we conclude that the apparent width w(l,s) depends linearly on s^2 with a different intercept and slope for each l. These functions can be summarized

by the equation

$$w(l,s) = w(l) - b(l)s^2,$$
 (22)



FIG. 7. Dependence of the film width on the magnetization at different values of *l*. Particle number in the simulation cell N = 1940.



FIG. 8. Top panel: dependence of the coefficient w(l) on the inverse value of l. Extrapolating to $l \to \infty$ we estimate the film width in the absence of the external magnetic field $w(l \to \infty) = w_0$. The red triangle indicates extrapolated value $w_0 = 4.46$. Bottom panel: dependence of the coefficient b(l) on the inverse value of l. Extrapolated to $l \to \infty$ value of this coefficient is related to the magnetostriction coefficient of the film by $b = b(l \to \infty) = \lambda w_0$. The red triangle indicates extrapolated value b = 0.03. Coefficients in Eq. (22) are estimated from the data shown in Fig. 7 for the film with particle number in the simulation cell N = 1940.

where $w(l) \equiv w(l, s = 0)$. Careful fitting indicates that b(l) has a systematic dependence on l. From this equation we can extract the l dependent magnetostriction coefficient as

$$\gamma(l) = \frac{w(l) - w(l,s)}{w(l)} = \lambda(l)s^2, \qquad (23)$$

where $\lambda(l) = b(l)/w(l)$. Finally, the magnetostriction coefficient for the films is defined as

$$\gamma = \gamma(l \to \infty) = \lambda s^2, \tag{24}$$

where $\lambda \equiv \lambda(l \to \infty)$. Repeating the procedure in films with a different number of particles *N* we find that λ has a dependence on *N* and we attempt next to determine the "best" value of λ . To this aim we fit the data in Fig. 7 and extrapolate the slopes and intercepts to $l \to \infty$. This is done in Fig. 8. It follows from Eq. (22), Eq. (23), and Eq. (24) that for a given film with *N* particles the film width at s = 0 is defined by $w(l \to \infty) = w_0(N)$ and the slope in Eq. (22) tends to $b(l \to \infty) = \lambda w_0(N)$.

In Fig. 9 we plot the simulations result for $b \equiv b(l \to \infty)$ as a function of w_0 as the black circles. The linear character of this



FIG. 9. The coefficient *b* as a function of the film width w_0 . Simulational results are represented by black dots. The theoretical results are the mean-field theory presented in Sec. VC culminating with Eq. (46).

relationship indicates that the saturation magnetostriction of wide enough films depends on their width as (see also Ref. [20])

$$\lambda = \lambda_{\text{bulk}} + \frac{\Lambda}{w_0},\tag{25}$$

where λ_{bulk} is the bulk value which is obtained when the width of the film tends to infinity. For purposes here we are interested in the slope of *b* vs w_0 (λ_{bulk}), not in the intercept (we will discuss the intercept below). Fitting a least-square line to the data we estimate

$$\lambda_{\text{bulk}} \approx (4.5 \pm 0.2) \times 10^{-3}.$$
 (26)

In the rest of this paper we provide the theory which will culminate in a first principle evaluation of this magnetostriction coefficient and we will also argue how we expect it to change with the width of the film.

V. CALCULATION OF THE MAGNETOSTRICTION

In this section we develop the general theory described in Sec. III with the aim of rationalizing the results obtained in the numerical simulations. Our aim is to compute $\langle \epsilon_{zz} \rangle$ as a function of *s*. As usual, since the strain tensor ϵ is not a state variable, we need to compute the stress response σ and extract the strain tensor from standard relations of elasticity theory. Thus our starting point is Eq. (17) for the response of the stress to the magnetic field. The RHS of this equation contains two terms, the Born term and the nonaffine fluctuations term.

A. The nonaffine term

To gain insight on the nonaffine term in Eq. (17) we employ our simulations. Measuring the two terms at temperature T = 0.23 we conclude that the two terms cancel each other and the stress and spin fluctuations are practically decoupled. In fact, dividing the difference between the two terms by the magnitude of either of them we find numbers of the order $10^{-5}-10^{-4}$ for all values of *m*. The same result was obtained for all the nonaffine components in the response of the tensor $\langle \sigma \rangle$. We thus conclude that to a high approximation

$$\langle \sigma_{\alpha\beta} m_{\gamma} \rangle \approx \langle m_{\gamma} \rangle \langle \sigma_{\alpha\beta} \rangle.$$
 (27)

It should be stated that in the ferromagnetic phase at low temperatures this conclusion may change drastically, and see for example the T = 0 results in Appendix B. Indeed, the athermal results in Refs. [4–7] show that nonaffine contributions to the magnetostriction and other responses are comparable in size to their respective Born terms. This can also be seen in the results quoted in Fig. 1 where the discontinuities in all the measured quantities are due to nonaffine responses.

A corollary of Eq. (27) is that in calculating the Born term in the response of the stress tensor we can employ a mean-field decoupling between the stress and the magnetization fluctuations. This simplifies the analytic calculation considerably.

B. Calculation of the Born term for magnetostriction

1. Definition of the strain tensor

In thermal systems the particles are restricted dynamically to their cages for long enough time τ which is nevertheless shorter than the diffusion time. Therefore we can compute a temperature dependent average position and an average spin orientation:

$$\langle \boldsymbol{r}_i \rangle = \frac{1}{\tau} \int_0^\tau dt \boldsymbol{r}_i(t), \quad \langle \boldsymbol{S}_i \rangle = \frac{1}{\tau} \int_0^\tau dt \boldsymbol{S}_i(t).$$
(28)

Once we apply an external strain $h_{ij}(\gamma)$ and a magnetic field **B** the average position and average spin orientation will experience an affine and a nonaffine response.

$$\langle \boldsymbol{r}_i \rangle(\boldsymbol{\gamma}, B) = h_{ij}(\boldsymbol{\gamma}) \langle \boldsymbol{r}_j \rangle + \boldsymbol{u}_i(\boldsymbol{\gamma}, B).$$
(29)

Here u_i is the nonaffine response that takes place as a result of the affine external strain and magnetic field, after which the system returns to thermal equilibrium. Defining the strain tensor $\epsilon_{\alpha\beta}$ in terms of the change in distance between pairs of particles,

$$\langle r_{ij} \rangle (\gamma, B) \equiv \langle r_{ij} \rangle \sqrt{1 + 2 \frac{\epsilon_{\alpha\beta} \langle r_{ij}^{\alpha} \rangle \langle r_{ij}^{\beta} \rangle}{\langle r_{ij} \rangle^2}},$$
 (30)

where r_{ij}^{α} is the α component of r_{ij} . Expanding this transformation to second order in $\epsilon_{\alpha\beta}$ we find,

$$\langle r_{ij} \rangle (\gamma, B) = \langle r_{ij} \rangle + \epsilon_{\alpha\beta} \frac{\langle r_{ij}^{\alpha} \rangle \langle r_{ij}^{\beta} \rangle}{\langle r_{ij} \rangle} - \frac{\epsilon_{\alpha\beta} \epsilon_{\gamma\delta} \langle r_{ij}^{\alpha} \rangle \langle r_{ij}^{\beta} \rangle \langle r_{ij}^{\gamma} \rangle \langle r_{ij}^{\delta} \rangle}{2 \langle r_{ij} \rangle^3}.$$
(31)

For the analysis below we define

$$\delta r_{ij} \equiv \langle r_{ij} \rangle (\gamma, B) - \langle r_{ij} \rangle. \tag{32}$$

2. The affine stress response

To compute $\langle \partial \sigma_{\alpha\beta} / \partial B_{\gamma} \rangle$ we express it in the form

$$\left\langle \frac{\partial \sigma_{\alpha\beta}}{\partial B_{\gamma}} \right\rangle = \frac{1}{V} \left\langle \frac{\partial^2 U}{\partial \epsilon_{\alpha\beta} \partial B_{\gamma}} \right\rangle. \tag{33}$$

Under a strain $\epsilon_{\alpha\beta}$ the mechanical energy (Lennard Jones interaction) will transform as:

$$U_{\text{mech}} = \sum_{i \neq j} \phi(r_{ij}) + \sum_{i \neq j} \frac{d\phi}{dr_{ij}} \delta r_{ij} + \frac{1}{2} \sum_{i \neq j} \frac{d^2\phi}{dr_{ij}^2} \delta r_{ij}^2.$$
(34)

Using Eq. (32) we can now write

$$U_{\text{mech}} = U_{\text{mech}}(\gamma = 0) + V \bigg[\epsilon_{\alpha\beta} c_{\alpha\beta}^{(1)} + \frac{1}{2} \epsilon_{\alpha\beta} \epsilon_{\gamma\delta} c_{\alpha\beta\gamma\delta}^{(2)} \bigg],$$
(35)

where

$$c_{\alpha\beta}^{(1)} = (1/V) \sum_{i \neq j} \frac{d\phi}{dr_{ij}} \frac{r_{ij}^{\alpha} r_{ij}^{\beta}}{r_{ij}}$$
$$c_{\alpha\beta\gamma\delta}^{(2)} = (1/V) \sum_{i\neq j} \left[\frac{1}{r_{ij}^{2}} \frac{d^{2}\phi}{dr_{ij}^{2}} - \frac{1}{r_{ij}^{3}} \frac{d\phi}{dr_{ij}} \right] r_{ij}^{\alpha} r_{ij}^{\beta} r_{ij}^{\gamma} r_{ij}^{\delta}.$$
 (36)

We can now express the mechanical contribution to the stress tensor $\sigma_{\alpha\beta}^{\text{mech}} = \partial \langle U_{\text{mech}} \rangle / V \partial \epsilon_{\alpha\beta}$ as

$$\sigma_{\alpha\beta} = c_{\alpha\beta}^{(1)} + \epsilon_{\gamma\delta} c_{\alpha\beta\gamma\delta}^{(2)} \quad \text{without exchange interaction.} \quad (37)$$

This is the stress in the material in the paramagnetic state in the absence of external magnetic fields. Let us now consider the additional strain imposed on the material in the ferromagnetic state or due to the application of an external magnetic field.

3. Exchange energy under strain in the glass phase

Under a strain $\epsilon_{\alpha\beta}$ the exchange coefficient $J(r_{ij})$ transforms as

$$J(\langle r_{ij} \rangle(\gamma)) = J(\langle r_{ij} \rangle) + \frac{dJ(r_{ij})}{dr_{ij}} \epsilon_{\alpha\beta} \frac{\langle r_{ij}^{\alpha} \rangle \langle r_{ij}^{\beta} \rangle}{\langle r_{ij} \rangle} + O(\epsilon^2),$$
(38)

where derivatives dJ(x)/dx are always computed at $x = \langle r_{ij} \rangle$. We have only expanded $J(r_{ij})$ to first order in the strain as in the exchange energy this will generate a term of $O(\epsilon s^2)$ and therefore the second order term can be neglected. The effect of strain $\epsilon_{\alpha\beta}$ on the average exchange interaction in the glass phase is then given by $\langle U_{ex} \rangle = -\sum_{i \neq j} J(r_{ij}) \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$. To estimate this term let us make the mean field approximation and replace $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle \approx \langle S_i \rangle \langle S_j \rangle = s^2$. Then we expand this energy to first order in $\epsilon_{\alpha\beta}$ as

$$\langle U_{\text{ex}} \rangle = -s^2 \left[\sum_{i \neq j} J(\langle r_{ij} \rangle) + \frac{dJ(r_{ij})}{dr_{ij}} \epsilon_{\alpha\beta} \frac{\langle r_{ij}^{\alpha} \rangle \langle r_{ij}^{\beta} \rangle}{\langle r_{ij} \rangle} \right], \quad (39)$$

or

$$\langle U_{\rm ex} \rangle = -V(a + \epsilon_{\alpha\beta} b_{\alpha\beta})s^2$$
 (40)

where we have used the notation

$$a = (1/V) \sum_{i \neq j} J(\langle r_{ij} \rangle)$$
$$b_{\alpha\beta} = (1/V) \sum_{i \neq j} \frac{dJ(r_{ij})}{dr_{ij}} \frac{r_{ij}^{\alpha} r_{ij}^{\beta}}{\langle r_{ij} \rangle}.$$
(41)

Combining the effects of strain on the mechanical and magnetic energies we find

$$\langle U \rangle = \langle U_{\text{mech}} + U_{\text{ex}} \rangle = U_{\text{mech}}(\gamma = 0) + V \left[\epsilon_{\alpha\beta} c_{\alpha\beta}^{(1)} + \frac{1}{2} \epsilon_{\alpha\beta} \epsilon_{\gamma\delta} c_{\alpha\beta\gamma\delta}^{(2)} - (a + \epsilon_{\gamma\delta} b_{\gamma\delta}) s^2 \right] (42)$$

So

$$\sigma_{\alpha\beta}(m) = \left\langle \frac{\partial U}{V \partial \epsilon_{\alpha\beta}} \right\rangle = c_{\alpha\beta}^{(1)} + \epsilon_{\gamma\delta} c_{\alpha\beta\gamma\delta}^{(2)} - b_{\alpha\beta} s^2.$$
(43)

C. Magnetostriction near $T = T_c$ for amorphous solids below the glass transition $T < T_g$

In our system, due to magnetization, a compressive strain is generated along the *z* direction. Further, the compressive stress obeys $\sigma_{zz} = 0$ as the film has a free surface both in the nonmagnetic and magnetic states. Thus in the nonmagnetic state we can use Eq. (35) to write

$$\sigma_{zz} = 0 = c_{zz}^{(1)} + \epsilon_{\gamma\delta} c_{zz\gamma\delta}^{(2)}, \qquad (44)$$

while in the magnetic state we can use Eq. (43) to write

$$\sigma_{zz}(m) = 0 = c_{zz}^{(1)} + \epsilon_{\gamma\delta} c_{zz\gamma\delta}^{(2)} + \gamma c_{zzzz}^{(2)} - b_{zz} s^2.$$
(45)

Subtracting Eq. (44) from Eq. (43) we find the magnetostriction coefficient

$$\gamma = \frac{b_{zz}s^2}{c_{zzzz}^{(2)}} = s^2 \frac{\sum_{i \neq j} \frac{1}{\langle r_{ij} \rangle} \frac{dJ(r_{ij})}{dr_{ij}} (z_i - z_j)^2}{\sum_{i \neq j} \left[\frac{1}{r_{ij}^2} \frac{d^2\phi}{dr_{ij}^2} - \frac{1}{r_{ij}^3} \frac{d\phi}{dr_{ij}}\right] (z_i - z_j)^4}.$$
 (46)

The first important result predicted by Eq. (46) is that the magnetostriction coefficient γ scales quadratically with the magnetization. The second important result concerns the width dependence of γ for a film of width w_0 . An accurate analysis of the results of the numerical simulations indicates that the saturation magnetostriction of wide enough films depends on the width according to Eq. (25). We can use our theory to understand this dependence. Using Eqs. (36) and (41), we can split formally the coefficient b_{zz} in Eq. (46) into two contributions $b_{zz} \approx b_{zz}^b + b_{zz}^s/w_0$ where the subscripts *b* and *s* stand for bulk and surface. Similarly we can write $c_{zzzz}^{(2)} \approx c_{zzzz}^{(2,b)} + c_{zzzz}^{(2,c)}/w_0$. Thus for magnetic films of width w_0 we find

$$\gamma(s, w_0) = s^2 \frac{w_0 b_{zz}^{\rm b} + b_{zz}^{\rm s}}{w_0 c_{zzzz}^{(2,b)} + c_{zzzz}^{(2,s)}}.$$
(47)

As w_0 tends to infinity we compute

$$\lambda_{\text{bulk}} = \frac{b_{zz}^{\text{b}}}{c_{zzz}^{(2,\text{b})}}.$$
(48)

In fact, we can use the general equation Eq. (46) to compute the magnetostriction coefficient of a film. In contrast to films on a substrate considered in Sec. IV this approach corresponds to films with two free boundaries. Nevertheless, we can compare the asymptotic bulk value of the saturated magnetostriction coefficient in these two cases. Plotting the coefficients of s^2 in Eq. (46) multiplied by w_0 (in order to obtain the coefficient *b*) as a function of w_0 we find the red triangles in Fig. 9. The best

linear fit results in the estimate

$$\lambda_{\text{bulk}} \approx 3.8 \times 10^{-3}. \tag{49}$$

Comparing with the numerical result in Eq. (26) we conclude that the agreement between the mean-field theory and the simulations is very satisfactory. Of course we need to stress that the theory still depends on the numerical measurement of the coefficients b_{zz} and c_{zzzz} .

Finally we can expand Eq. (47) in inverse powers of w_0 and the leading result will read exactly like Eq. (25). The coefficient Λ cannot be directly compared between theory and experiment because it stems from two different sources. One is purely geometric, particles in the center of the film have more interaction than close to the two surfaces. The second comes from the interaction between the film and substrate. In the theory we did not take particular care of the interaction between the film and the substrate so the intercept in Fig. 9.

VI. SUMMARY AND DISCUSSION

In summary, we have presented a theory for the mechanical and magnetic responses of amorphous solids which is equally applicable to a bulk sample or a film whose width is in the nano scale. In this paper we focused on the magnetostriction as a good measure of the interplay between mechanical strain and magnetic fields. Analytic theory for all the other responses was offered both at T = 0 or at finite temperature. We found that for intermediate temperatures between T_c and T_g the nonaffine contribution to the magnetostriction was negligible in the nanofilm. This simplifies the theoretical calculation of the magnetostriction coefficient which is found to be in good agreement with the numerical simulations. Interestingly enough, both in bulk and in nanofilm the magnetostriction coefficient is proportional to s^2 and therefore to the square of the magnetization. It is expected that at low temperatures, 0 < $T \ll T_c$, the nonaffine contribution should be significant, since at T = 0 it is of the same order as the Born contribution. It is therefore interesting to examine this issue both in experiments and in simulations at this range of temperatures.

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APPENDIX A: MONTE CARLO EQUILIBRATION

The Monte Carlo (MC) simulations were performed in both NPT (in a bulk phase) and NVT (in a film) ensembles. In a bulk phase we start with an initial face-centered-cubic arrangement of A type particles with periodic boundary conditions in three directions. Then a randomly chosen 20% of the particles were changed to B type. The initial configuration of a film (of desired height) was cut along the z axis from a bulk glass and put on top of the crystalline substrate. Then the system (the bulk phase or the film) was equilibrated at high temperature T = 5. Once equilibrated, the systems were quenched instantaneously to T = 0.23. In each NVT ensemble MC sweep we attempt to move each particle once. We chose the maximum position displacement such that the acceptance ratio of the trial moves was

around 30%. In the case of NPT ensembles, in addition to the trial moves we attempt to change the length of the simulation box in every 20 MC sweeps. We chose the maximum change in box length such that the acceptance ratio of the trial moves was around 30%. Optimum particle displacements and changes in the box length are obtained for 200000 MC sweeps before starting to gather thermal statistics. To update the spins, we use Wolf's cluster algorithm [21] when there is no external magnetic field. We made two modifications to this algorithm. Firstly, concentrating on any given particle *i* we refer to its neighbors, as any particle *j* that resides within a distance of 2.5 from it. Secondly, the coupling defined by $J(r_{ii})$ [see Eq. (9)] is not a constant as in a common lattice problem. We attempt the particle move and spin flip in the following sequence: Two sweeps, in each of which we attempt to move each particle once, are followed by the construction of one Wolf cluster after which the Monte Carlo proceeds with the next two sweeps. In the presence of magnetic field the Wolf's cluster algorithm is not effective. Hence we apply a single spin flip algorithm in which we attempt to randomly flip each spin once.

APPENDIX B: RESPONSES AT ZERO TEMPERATURE

All the important response functions exhibited by magnetic amorphous solids at T = 0 have been studied in great detail [16,22,23] and can be expressed in terms of the eigenvalues and eigenfunctions of a Hessian matrix H for N particles in d dimensions where

$$H_{ij}^{(rr)} \equiv \frac{\partial^2 U}{\partial r_i \partial r_j} \qquad (dN \times dN \text{ matrix})$$

$$H_{ij}^{(rS)} \equiv \frac{\partial^2 U}{\partial r_i \partial S_j} \qquad (dN \times dN \text{ matrix})$$

$$H_{ij}^{(Sr)} \equiv \frac{\partial^2 U}{\partial S_i \partial r_j} \qquad (dN \times dN \text{ matrix})$$

$$H_{ij}^{(SS)} \equiv \frac{\partial^2 U}{\partial S_i \partial S_j} \qquad (dN \times dN \text{ matrix}),$$
(B1)

and four 'mismatch forces' Ξ that represent the forces and torques on the particles before the nonaffine flows ensure new local minima for the particle positions and spins. For notational simplicity let us assume that we can replace the stress tensor $\epsilon_{\alpha\beta}$ by a scalar γ here, then the particle positions and spins can be written $\{r_i(\gamma, B)\}, \{S_i(\gamma, B)\}$. Then the mismatch forces can be written

$$\Xi_{i}^{(\gamma,r)} \equiv \frac{\partial^{2}U}{\partial\gamma\partial r_{i}}
\Xi_{i}^{(\gamma,S)} \equiv \frac{\partial^{2}U}{\partial\gamma\partial S_{i}}
\Xi_{i}^{(B,r)} \equiv \frac{\partial^{2}U}{\partial B\partial r_{i}}
\Xi_{i}^{(B,S)} \equiv \frac{\partial^{2}U}{\partial B\partial S_{i}}.$$
(B2)

In terms of these Hessian and mismatch forces we can find all the modulii that describe the mechanical and magnetic properties of magnetic glasses. Thus the shear modulus takes the form

$$\mu(\gamma, B) = \frac{\partial^2 U}{\partial \gamma^2} \Big|_B - \Xi^{(\gamma)} \cdot \boldsymbol{H}^{-1} \cdot \Xi^{(\gamma)}.$$
(B3)

Note the nonaffine contribution reduces the shear modulus.

The magnetic susceptibility $\chi(\gamma, B)$ can similarly be expressed in terms of a classic thermodynamic form that exists for crystalline solids and an additional term required for magnetic equilibrium in the case of amorphous solids

$$\chi(\gamma, B) = -\frac{\partial^2 U}{\partial B^2}\Big|_{\gamma} + \Xi^{(B)} \cdot \boldsymbol{H}^{-1} \cdot \Xi^{(B)}.$$
(B4)

Here the additional positive definite form exists due to the existence of nonaffine flows that can help minimize the potential energy of the magnetic glass. Magnetostriction can be measured from the change of stress of a specimen with changing magnetic field B

$$\chi_{\sigma,B}(\gamma,B) = \frac{d\sigma}{dB}\Big|_{\gamma} = \frac{\partial^2 U}{\partial B \partial \gamma} - \Xi^{(\gamma)} \cdot H^{-1} \cdot \Xi^{(B)}.$$
 (B5)

While magnetoelasticity and magnetoplasticity involve the magnetic response of a material to applied strain

$$\chi_{M,\gamma}(\gamma,B) = \frac{dM}{d\gamma}\Big|_{B} = -\frac{\partial^{2}U}{\partial\gamma\partial B} + \Xi^{(B)} \cdot H^{-1} \cdot \Xi^{(\gamma)}.$$
(B6)

Note that for crystalline solids we have the Maxwell relation between magnetostriction and magnetoelasticity $\frac{\partial \sigma}{\partial B}|_{\gamma} = -\frac{\partial M}{\partial \gamma}|_{B}$. For metallic glasses as the Hessian matrix is hermitian we have an analogous Maxwell relationship

$$\left. \frac{d\sigma}{dB} \right|_{\gamma} = -\frac{dM}{d\gamma} \right|_{B}.$$

- [1] J. D. Livingston, Phys. Stat. Sol. 70, 591 (1982).
- [2] G. During and S. Zapperi, J. Appl. Phys. 85, 5196 (1999).
- [3] L. Santi, F. Bohn, A. D. C. Viegas, G. Durin, A. Magni, R. Bonin, S. Zapperi, and R. L. Sommer, Physica B: Condensed Matter 384, 144 (2006).
- [4] H. G. E. Hentschel, V. Ilyin, and I. Procaccia, Europhys. Lett. 99, 26003 (2012).
- [5] R. Dasgupta, H. G. E. Hentschel, I. Procaccia, and B. S. Gupta, Europhys. Lett. 104, 47003 (2013).
- [6] H. G. E. Hentschel, I. Procaccia, and B. S. Gupta, Europhys. Lett. 105, 37006 (2014).
- [7] H. G. E. Hentschel, V. Iliyn, I. Procaccia, and B. S. Gupta, J. Stat. Mech. (2014) P08020.
- [8] A. K. Dubey, H. G. E. Hentschel, P. K. Jaiswal, C. Mondal, I. Procaccia, B. S. Gupta, Europhys. Lett. 112, 17011 (2015).
- [9] H. G. E. Hentschel, I. Procaccia and B. S. Gupta, Phys. Rev. E 93, 033004 (2016).
- [10] N. Akulov, Z. Phys. 52, 389 (1928).
- [11] W. Kob and H. C. Andersen, Phys. Rev. E 51, 4626 (1995).

- [12] K. Vollmayr, W. Kob, and K. Binder, J. Chem. Phys. 105, 4714 (1996).
- [13] S. Sastry, P. G. Debenedetti, and F. H. Stillinger, Nature (London) 393, 554 (1998).
- [14] E. Lomba, J. L. López-Martín, H. M. Cataldo, and C. F. Tejero, Phys. Rev. E 49, 5164 (1994).
- [15] J. P. Wittmer, H. Xu, P. Polinska, F. Weysser, and J. Baschnagel, J. Chem. Phys. 138, 12A533 (2013).
- [16] A. Lemaitre and C. Maloney, J. Stat. Phys. 123, 415 (2006).
- [17] B. E. Lorenz and C. D. Grahem, Jr., IEEE Trans. Magn. 42, 3886 (2006).
- [18] B. D. Cullity and C. D. Grahem, *Introduction to Magnetic Materials* (Wiley-IEEE Press, Hoboken, New Jersey, 2009).
- [19] A. Haji-Akbari and P. Debenedetti, J. Chem. Phys. 141, 024506 (2014).
- [20] T. Szumiata, M. Gzik-Szumiata, and K. Brzózka, Mater. Sci.-Poland 26, 1039 (2008).
- [21] U. Wolf, Phys. Rev. Lett. 62, 361 (1989).
- [22] J. F. Lutsko, J. Appl. Phys. 64, 1152 (1988).
- [23] J. F. Lutsko, J. Appl. Phys. 65, 2991 (1989).