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Letter to the Editor

The reversible transverse susceptibility of coupled uniaxial systems

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Abstract

The linear response of two interacting uniaxial systems is studied and the reversible transverse susceptibility (RTS) is calculated. There exist up to four nucleation fields at which RTS becomes singular. However, we find that nucleation of highly symmetric extremal energy states (identical components parallel or antiparallel to applied field) is not accompanied by a singularity in RTS. Specific examples are given and the case of $n > 2$ interacting components is discussed.

The reversible transverse susceptibility (RTS) is well known to have singularities (peaks) at the nucleation fields of the observed system. Recently, Hoare et al. [1] presented experimental measurements of RTS in particulate recording media together with an analysis based on a theory developed by Aharoni et al. [2] for noninteracting uniaxial particles with coherent rotation of magnetization. More complicated symmetries have been analyzed in great detail by Asti et al. [3 and references therein] who considered also nonlinear response. The suppression and shift of RTS

peaks due to thermal relaxation effects in uniaxial systems was discussed by Lu et al. [4].

As opposed to the exhaustively studied noninteracting magnetic systems very few analytic results are available for interacting systems, a recent notable exception is the work of Chen et al. [5] on a pair of uniaxial particles with dipole–dipole coupling. Here we present, in brief, a general discussion of linear response to an infinitesimal external force field and apply this theory to a binary coupled system with energy E . Similar to Asti et al. [3] we find that high symmetry of the multidimensional configuration space suppresses possible singularities in RTS.

Let now $E = E(x_i | H)$ with x_i being the internal degrees of freedom of the system about to be

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perturbed and H the applied field. At a local extremum $\bar{x}_i$, where $\partial E/\partial x_i = 0$ for all i , we define the Hessian of the energy function by the determinant

$$\Delta(\bar{x}_i) = \det \left(\frac{\partial^2 E}{\partial x_m \partial x_n} \right) \bigg|_{\bar{x}_i}. \quad (1)$$

The vanishing of the Hessian (1) is a necessary, but *not* sufficient, condition that RTS has a singularity at the corresponding critical field. We shall call critical field, H_c , any external applied field H for which the Hessian (1) vanishes at any of the local energy extrema. In contrast to the decoupled case not every critical field H_c is a nucleation field H_n at which the system E changes its degree of multistability, i.e. at which a local minimum is either nucleated or destroyed. The total energy of the system is $E_{\text{tot}} = E(x_i | H) + h\epsilon(x_i)$ where $E(x_i | H)$ is the unperturbed energy and h is an infinitesimal perturbing field. The extremal points $\bar{x}_i(h)$ satisfy the conditions $\partial E_{\text{tot}}/\partial x_i |_{\bar{x}_i} = 0$ and we shall seek the functions $\bar{x}_i(h)$ to the first order in h . To this end we write $\bar{x}_i(h) = \bar{x}_i(0) + h\delta\bar{x}_i + o(h)$

and demand that the k extremal conditions

$$0 = \frac{\partial E}{\partial x_k} + h \left[\frac{\partial \epsilon}{\partial x_k} + \sum_l \delta\bar{x}_l \frac{\partial^2 E}{\partial x_k \partial x_l} \right] + o(h) \quad (2)$$

be satisfied separately in every order of h . The derivatives of E are to be calculated at the unperturbed extrema $\bar{x}_i(0)$ and the first order term constitutes a set of linear equations for the perturbation $\delta\bar{x}_i$; its discriminant is the Hessian (1) so that $\delta\bar{x}_i \rightarrow \infty$ as $\Delta(\bar{x}_i) \rightarrow 0$, i.e. at any critical field H_c . However, it may happen that not all of the equations (2) are linearly independent and in this case the perturbations $\delta\bar{x}_i$ remain finite even as $\Delta \rightarrow 0$. We show below that this happens on nucleation of a highly symmetric local minimum.

Assuming coherent rotation we apply Eq. (2) to two coupled uniaxial components with volumes V_i , anisotropy constants $K_i > 0$ and saturation magnetizations $M_i = |\mathbf{M}_i|$. Their easy axes are in the $[\sin \beta_i, 0, \cos \beta_i]$ directions and both are driven by two periodic biasing fields $\mathbf{H}(t)$ and $\mathbf{h}(t)$ applied perpendicular to each other:

$$\mathbf{H} = H(t)[0, 0, 1] \text{ and } \mathbf{h} = h(t)[\cos \gamma, \sin \gamma, 0].$$

The field $H(t)$ varies slowly and drives a hysteresis loop while the perturbing field $h(t)$ varies rapidly with infinitesimal amplitude $h \rightarrow 0$. The total energy of the coupled system is

$$E_{\text{tot}}(\theta_i, \phi_i | H, h) = \sum_{i=1}^2 (E_i + h\epsilon_i) + B(\mathbf{M}_1 \cdot \mathbf{M}_2) + \mathbf{M}_1 \cdot \mathbf{C} \cdot \mathbf{M}_2 \quad (3)$$

where $\mathbf{C}$ is a symmetric tensor. The single component energies are

$$E_i(\theta_i, \phi_i | H) = -K_i V_i (\sin \beta_i \cos \phi_i \sin \theta_i + \cos \beta_i \cos \theta_i)^2 - V_i \mathbf{M}_i \cdot \mathbf{H} \quad (4)$$

and $\epsilon_i = -V_i \mathbf{M}_i \cdot \mathbf{h}/h$ is the perturbative term. In the spherical coordinates $\theta_i \in \langle 0, \pi \rangle$, $\phi_i \in \langle 0, 2\pi \rangle$, the magnetization vectors become

$$\mathbf{M}_i = M_i [\sin \theta_i \cos \phi_i, \sin \theta_i \sin \phi_i, \cos \theta_i].$$

We also set $k_i = 2K_i V_i$ and $m_i = M_i V_i$ for brevity.

Equations (3) and (4) find application in the theory of magnetic bilayers [6], of noninteracting coated particles [7] where the coating acts as a magnetic system coupled to the particle's core, of two-sublattice systems [8] and of two particles with dipole–dipole coupling [5]. However, they are still too complicated for analysis and we shall impose yet an additional constraint on their geometry, to wit, that all extrema of the unperturbed energy lie along the $\phi_i = 0$ and π axes. Their location may then be found from the function $E(\theta_i) \equiv E(\theta_i, 0)$ in which the domain of the angles θ_i has been extended to the toroidal surface $\langle 0, 2\pi \rangle \times \langle 0, 2\pi \rangle$. The mixed derivatives $\partial^2 E/\partial \theta_i \partial \phi_j$, $i, j = 1, 2$, are then all identically zero at the local extrema and the linear set (2) takes on a block form. In full analogy to the noninteracting case [2] the perturbations $\delta\phi_i$ remain finite (in this geometry only) and we shall concentrate on the shifts $\delta\theta_i$ found from the linear set

$$\sum_j w_{ij} \delta\theta_j = m_i \cos \theta_i \cos \gamma, \quad (5)$$

where $w_{ij}(H) = \partial^2 E/\partial \theta_i \partial \theta_j$ evaluated at a local minimum. The discriminant of Eqs. (5), $\Delta(H) = w_{11}w_{22} - w_{12}^2$, is the Hessian (1) and the perturbations

$$\delta\theta_1 = \cos \gamma (m_1 \cos \theta_1 w_{22} - m_2 \cos \theta_2 w_{12})/\Delta \quad (6)$$

and $\delta\theta_2$, obtained by letting $1 \leftrightarrow 2$, tend to infinity if $\Delta \rightarrow 0$, i.e. at the critical points of the system (3). There is, however, an exception: If $m_1 = m_2$, $\theta_1 = \theta_2$ and $w_{11} = w_{22}$ (identical particles) then

$$\delta\theta_1/m_1 = \cos \gamma \cos \theta_1 / (w_{22} + w_{12}) = \delta\theta_2/m_2$$

and the perturbations are regular.

A binary system has upto four local minima at $(\theta_i^{(k)}, \phi_i^{(k)})$ and the projection of its total magnetic moment $\mathbf{m}$ into the direction of $\mathbf{h}$ is

$$m_h = \sum_{k=1}^4 n^{(k)} \sum_{i=1}^2 m_i \sin \theta_i^{(k)} \cos(\phi_i^{(k)} - \gamma). \quad (7)$$

The numbers $n^{(j)} \geq 0$, such that $\sum_j n^{(j)} = 1$, give the immediate state of the system (3) under the action of the field $H(t)$. Their interpretation depends on the nature of this system: Measurements are, as a rule, carried out on one bilayer in a definite configuration at one of the local energy minima. Thus only one of the numbers $n^{(j)}$ can be nonzero at a time and, as the field $H(t)$ is varied, they switch between 1 and 0 at the nucleation fields H_n . This Stoner–Wohlfarth picture [9] is often used also in particulate media where the response of a large assembly of particles is being measured. If, however, random switching events due to thermal relaxation are here taken into account then $n^{(j)}(t)$ become continuous functions of time denoting [4,10] the reduced mean number of particles magnetized in one of the possible easy directions. From Eq. (7) we finally define RTS as

$$\chi_t = \lim_{h \rightarrow 0} \partial M_h / \partial h$$

where $M_h = m_h / (V_1 + V_2)$ is the projection of magnetization into the direction of the field $\mathbf{h}$. Setting $n^{(j)} \rightarrow n^{(j)} + h \delta n^{(j)}$ we obtain

$$(V_1 + V_2) \chi_t = \sum_j \sum_{i=1}^2 m_i \cos \theta_i^{(j)} \delta \phi_i^{(j)} n^{(j)}, \quad (8)$$

where we omitted the regular $\delta \phi_i^{(j)}$ terms (this is exact if $\gamma = 0$) and the equally regular incoherent $\delta n^{(j)}$ terms [4] which depend on the detailed dynamics of the system. The shifts $\delta \phi_i^{(j)}$, required in treatment of disordered samples [1,4], are easily calculated from the general formula (2). Note

that the singularities of $\delta \theta_i^{(j)}$ are observable only if the occupation number $n^{(j)}$ is nonzero, i.e. if the vanishing local minimum state is not empty.

Even in our chosen simple geometry it is not possible, in general, to find analytically the local minima and we shall restrict our analysis to two special cases, the first of them being a two particle cluster with dipole–dipole interactions [5] so that $B = 0$. The particles are assumed to be identical, $K_i = K$, $V_i = V$ and $M_i = M$, and aligned with the applied field H , $\beta_i = 0$. Their separation is r , and thus $r^3 C_{11} = 1 - 3 \sin^2 \beta_p$, $r^3 C_{13} = -3 \sin \beta_p \cos \beta_p$ and $r^3 C_{33} = 1 - 3 \cos^2 \beta_p$ where β_p is the bond angle. For simplicity we also set $\gamma = 0$. Following Chen et al. [5] we introduce the reduced applied field $b = H/H_n$ ($H_n = 2K/M$ is the nucleation field of an isolated particle) and the coupling constant $\rho = M^2 V / (2K r^3)$ and restrict ourselves to the two values of the bond angle $\beta_p = 0$ and π . In either case there exist upto three physically distinct configurations corresponding to a local energy minimum: $\uparrow\uparrow$ at $(0, 0)$, the equivalent $\uparrow\downarrow$ and $\downarrow\uparrow$ states at $(0, \pi)$ and $(\pi, 0)$ and $\downarrow\downarrow$ at (π, π) .

The case of bond angle $\beta_p = 0$ shall be considered first.

The Hessians at the possible locations of local minima become

$$\begin{aligned} \Delta(0, 0|b) &= \Delta(\pi, \pi| -b) \\ &= 4K^2 V^2 (b_0 + b)(b_1 + b), \\ \Delta(0, \pi|b) &= \Delta(\pi, 0|b) = 4K^2 V^2 (b_2^2 - b^2), \end{aligned} \quad (9)$$

(10)

where the three reduced critical fields $b_0 = 1 + 3\rho$, $b_1 = 1 + \rho$ and $b_2 = (1 - \rho)^{1/2}(1 - 3\rho)^{1/2}$ satisfy the inequalities $b_0 \geq b_1 \geq b_2$. The system is tristable [5] at fields $|b| < b_2$, bistable at $b_2 \leq |b| \leq b_1$ and monostable for $|b| > b_1$. The point $(0, 0)$ is a saddle on the field interval $-b_0 < b < -b_1$ and an absolute maximum for $b < -b_0$; b_0 is thus not a nucleation field since a saddle point cannot be occupied.

We encounter here the exceptional suppression of the RTS peak discussed at Eq. (6): There is $w_{11} = w_{22}$ at $(0, 0)$ and

$$\delta\theta_1 = \delta\theta_2 = H_n^{-1} (b_0 + b)^{-1}, \quad (11)$$

but the state $(0, 0)$ must necessarily be empty at fields $|b| \geq b_1$ and by Eqs. (7) and (8) the singularity of $\delta\theta_1$ at $b = -b_0$ is by definition not observable. Identical considerations, with $b \rightarrow -b$, hold also for the symmetric (π, π) state. The intermediate states $\uparrow\downarrow$ and $\downarrow\uparrow$ at $(0, \pi)$ and $(\pi, 0)$ may be occupied (depending on the dynamics, see below) for fields $|b| < b_2$ and the corresponding shifts are

$$\delta\theta_i = H_n^{-1} [b \mp (1 - 3\rho)] (b_2^2 - b^2)^{-1} \quad (12)$$

(upper sign corresponds to $i = 1$). In the limit of zero interaction strength, $\rho \rightarrow 0$, both Eq. (11) and (12) go over to the single particle result [2]. The peak (11) corresponding to nucleation of the symmetric states $\uparrow\uparrow$ and $\downarrow\downarrow$ is being progressively suppressed with increasing interaction strength as the difference $b_0 - b_1 = 2\rho$ grows. For $\rho > 1/3$ the nucleation field b_2 is no longer defined, the asymmetric states do not exist, and, by virtue of Eq. (8) only the regular functions (11) contribute to the RTS field dependence which thus becomes totally free of singularities.

Given the barrier heights [5] separating the local minima it is easy to set up a master equation [4,10] for the occupation numbers $n^{(j)}$ and to calculate the hysteresis loop of the system (3) taking into account thermally driven transitions between the three levels. We have recently solved [11] these equations and found that the intermediate states $\uparrow\downarrow$ and $\downarrow\uparrow$ are almost always empty. One exception is the case of very slow sweeps of the field $H(t)$ where the system remains close to thermal equilibrium (but this is relevant only at very high temperatures) and the other exception is the Stoner–Wohlfarth limit [9] (fast sweeps and low temperatures). Only in the latter case have the intermediate states a significant population close to the nucleation field b_2 , i.e. close to the singularities of Eq. (12). It may also be noted here that the prefactor of thermal relaxation rate has a strong field dependence and rapidly vanishes as $H \rightarrow H_n$ [12]. Thermal relaxation is thus slowed down and the Stoner–Wohlfarth limit is reached (at a given sweep rate) at higher temperatures than predicted by theory with constant prefactor [4,10].

The case of bond angle $\beta_p = \pi/2$ is fully analogous: The Hessians (9) and (10) retain their form, with critical fields $b_0 = 1 + \rho$, $b_1 = 1 - 3\rho$ and $b_2 = (1 - \rho)^{1/2}(1 + 3\rho)^{1/2}$ and inequalities $b_0 \geq b_2 \geq b_1$. The system is tristable [5] at fields $|b| < b_1$, bistable at $b_1 \leq |b| \leq b_2$ (but it is the symmetric states $\uparrow\uparrow$ and $\downarrow\downarrow$ which collapse at $b = \pm b_1$) and monostable for $|b| > b_1$ where the asymmetric states vanish; b_0 is again not a nucleation field. The response of the symmetric states is finite, given by Eq. (11), while the response of the asymmetric states diverges according to Eq. (12) with $\rho \rightarrow -\rho$. Model calculations of the hysteresis loop show that the intermediate asymmetric states are again almost always empty.

We shall now use a simple bilayer model to study the linear response of a coupled binary system with nonidentical components. In Eq. (3) we write to this end $\mathbf{M}_1 \cdot \mathbf{C} \cdot \mathbf{M}_2 = 2C\mathbf{M}_1 \cdot \mathbf{M}_2$ where C is a number. There exist four nonequivalent local minima at $(0, 0)$, $(0, \pi)$, $(\pi, 0)$ and (π, π) , the corresponding Hessians being

$$\begin{aligned} \Delta(0, 0|H) &= \Delta(\pi, \pi| -H) \\ &= m_1 m_2 (H + H_1)(H + H_2), \end{aligned} \quad (13)$$

$$\begin{aligned} \Delta(0, \pi|H) &= \Delta(\pi, 0| -H) \\ &= -m_1 m_2 (H + \bar{H}_1)(H + \bar{H}_2). \end{aligned} \quad (14)$$

Let $\rho_1 = 2(B + C)$ and $\rho_2 = 2(B - C)$ be two reduced coupling constants. The critical fields

$$\begin{aligned} 2m_1 m_2 H_i &= k_1 m_2 + k_2 m_1 - \rho_1(m_1 + m_2) \\ &+ (-1)^i D, \end{aligned} \quad (15)$$

$$\begin{aligned} D^2 &= [k_1 m_2 - k_2 m_1 + \rho_1(m_1 - m_2)]^2 \\ &+ 4\rho_1^2 m_1 m_2, \end{aligned}$$

go, in the decoupled limit, to the nucleation fields of individual components, $H_i \rightarrow k_i/m_i = 2K_i/M_i$. In the presence of interactions, however, only H_1 (or, rather, $-H_1$) is a nucleation field at which the local minimum at $(0, 0)$ turns into a saddle point. This by definition unoccupied state be-

comes a local maximum for fields $H < -H_2$. Similarly, the two critical fields

$$2m_1m_2\bar{H}_i = \bar{D} + (-1)^i \times \{k_2m_1 - k_1m_2 - \rho_2(m_1 - m_2)\}, \quad (16)$$

$$\bar{D}^2 = [k_1m_2 + k_2m_1 - \rho_2(m_1 + m_2)]^2 - 4\rho_2^2m_1m_2,$$

go over to k_i/m_i in the decoupled limit, but, contrary to the previous case, they both represent a nucleation field.

Considering now the perturbations $\delta\theta_i$ of Eq. (6) we note that

$$w_{ii}(0, 0|H) = w_{ii}(\pi, \pi|-H) = k_i + m_iH - \rho_1,$$

$$w_{ii}(0, \pi|H) = w_{ii}(\pi, 0|-H)$$

$$= k_i - (-1)^i m_iH - \rho_2,$$

$w_{12} = 2\rho_1$ at $(0, 0)$ and (π, π) and $w_{12} = 2\rho_2$ at $(0, \pi)$ and $(\pi, 0)$ whence the shifts $\delta\theta_i$ are easily calculated. In the limit of two identical components $H_1 < H_2$ in Eq. (15) but $\bar{H}_1 = \bar{H}_2$ in Eq. (16) so that Eqs. (11) and (12) are recovered.

In the general case of misaligned components of the system (3) there exist upto four distinct nucleation fields since the states $\uparrow\uparrow$ and $\downarrow\downarrow$ become asymmetric.

In summary, we find that the linear response of a binary interacting system is governed by the system symmetry, a fact pointed out previously for noninteracting systems by Asti et al. [3]. At the nucleation field H_n of a general minimum energy configuration the RTS diverges as $\chi_i \propto (H - H_n)^{-1}$ in full analogy to the noninteracting case. Exceptional is the case of identical components: If there exists a fully symmetric configuration of minimum energy, i.e. if locally $E(\theta_1, \theta_2|H) = E(\theta_2, \theta_1|H)$, then linear response remains finite at the nucleation field of this state.

It would be highly desirable to generalize the above result to $n > 2$ interacting particles. In the fully symmetric state $w_{ii} = w$ and $w_{ij} = \bar{w}$ for $i, j = 1, \dots, n$ and the corresponding Hessian is $\Delta = w^2 - \bar{w}^n$. It is easy to show that the solutions of

Eq. (5), with all m_i and θ_i identical, remain finite as $w - \bar{w} = 0$ but it is not clear (to us) how to prove that this equation determines the *unique* nucleation field. Geometric intuition suggests this to be indeed true since there should exist only one field at which a local minimum unfolds into the degenerate saddle point which is not a local maximum along any curve of the multidimensional space. Further action of the field transforms the saddle in manifold ways (corresponding to the roots of $\Delta(w - \bar{w})^{-1} = 0$) till finally it becomes a local energy maximum.

The above arguments constitute by no means a proof but we do propose that the linear response of highly ordered arrays of identical interacting components remains finite as fully symmetric ground states (all up or all down) are nucleated or annihilated.

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