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# On the magneto-elastic properties of elastomer–ferromagnet composites

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## Abstract

We study the macroscopic magneto-mechanical behavior of composite materials consisting of a random, statistically homogeneous distribution of ferromagnetic, rigid inclusions embedded firmly in a non-magnetic elastic matrix. Specifically, for given applied elastic and magnetic fields, we calculate the overall deformation and stress–strain relation for such a composite, correct to second order in the particle volume fraction. Our solution accounts for the fully coupled magneto-elastic interactions; the distribution of magnetization in the composite is calculated from the basic minimum energy principle of magneto-elasticity. © 2001 Elsevier Science Ltd. All rights reserved.

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## 1. Introduction

We study the macroscopic magneto-mechanical behavior of composite materials consisting of a distribution of magnetically permeable rigid inclusions embedded firmly in a non-magnetic elastic matrix. By analogy with their fluid counterparts—magneto- and electro-rheological fluids—such composites are generally called magneto-rheological (MR) solids. The inclusions in MR solids are typically micron sized particles of iron or iron-based alloys such as carbonyl–iron and iron–cobalt (Jolly et al., 1996a; Rigbi and Jilken, 1983); the non-magnetic matrix, in turn, is an elastomer like rubber,

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a polymer gel, etc. In this paper, we evaluate the overall magneto-elastic response of MR solids *to second order* in the particle volume fraction  $\Phi$ —that is, to the lowest order in the volume fraction expansion for which the magnetic interactions between particles are taken into account.

Upon application of a constant magnetic field  $\mathbf{H}_0$ , the overall shape and elastic properties of a MR solid are altered rapidly and reversibly. Of course, the mechanism responsible for this bulk effect is the induced magnetic interaction between the ferromagnetic particles in the composite. Most theoretical studies of MR solids assume microgeometries consisting of chains of particles. Such geometries, which occur naturally in MR fluids, can also be induced in MR solids through application of strong magnetic fields during the elastomer crosslinking process. Indeed, application of a strong magnetic field at the crosslinking stage gives rise to particle alignment and formation of chains which are then locked in place upon final cure (Jolly et al., 1996a, b; Rigbi and Jilken, 1983). Based on the assumption of a microgeometry in which particles are arranged in chains, some of the existing theoretical studies (Jolly et al., 1996a) account for magnetic dipole interactions between adjacent particles in a chain: dipoles are considered as would be induced by the applied magnetic field in an isolated sphere and the forces between two such dipoles are then evaluated—and used to obtain the overall response of the composite. Other studies of chain geometries (Bonnetcaze and Brady, 1992; Ginder and Davis, 1994) resort to numerical calculations of the magnetic part of the problem together with empirical expressions for the non-linear magnetization, and utilize geometrical approximations under which the magnetic and the elastic problems decouple.

In this paper, we consider the fully coupled magneto-elastic problem in MR solids for random microgeometries, and we evaluate the overall properties of MR solids in the important regime in which the volume fraction  $\Phi$  of particles is small. Our calculations, which provide an expansion of the overall properties *to second order in the volume fraction*, account correctly for interparticle magneto-elastic interactions. Further, the present framework does not utilize empirical magnetization expressions; instead, magnetizations are obtained from the basic principles of minimum magneto-elastic energy (Brown, 1962). Our calculations assume that the ferromagnetic particles are uniformly magnetized. Such an assumption is justified if the particles are small enough ( $\leq 1.5 \mu\text{m}$  for iron) in such a way that magnetic microgeometries consisting in more than one laminate are thermodynamically unfavorable (Landau and Lifshitz, 1996; Morish, 1965). Further, our calculations neglect anisotropy effects in the magnetization. This approximation is justified for cubic crystalline ferromagnetic materials, such as iron and iron–cobalt alloys (Landau and Lifshitz, 1996) and for polycrystalline particles. For ferromagnetic particles that satisfy the above assumptions, our method of solution is almost entirely analytical. For other cases, both the anisotropy effects and the existence of varying magnetizations within the particles must be taken into account. These complications can only be dealt with numerically. However, a treatment of the general case can be built upon the method of solution for the simpler case presented in this paper.

Our analysis applies to composites consisting of random, statistically homogeneous distributions of ferromagnetic inclusions within an elastic matrix; our results give the

overall deformations and stress–strain relation for such a composite under given applied elastic and magnetic fields. To study the effect of magnetic interactions in the composite on its bulk properties, we calculate the average strain for various applied tractions and magnetic fields. In order to take advantage of known elasticity solutions for systems consisting of pairs of inclusions in an elastic matrix—in the present problems which, as we will see, can be dealt with more easily by prescribing displacements—we introduce a novel procedure, which calls for minimization of a certain energy expression over a finite dimensional space of homogeneous strains.

Our examples show that the response of MR solids containing randomly distributed particles depends strongly on the applied magnetic field  $\mathbf{H}_0$ . Qualitatively, the magnetostatic interactions associated with the random distribution of inclusions induce forces that oppose deformations which tend to lengthen the material in the direction parallel to the applied magnetic field. Further, the sole application of a magnetic field can induce a substantial deformation in the composite. This deformation consists of an overall compression, although the strain in the direction of the applied magnetic field is different from the strain in directions orthogonal to  $\mathbf{H}_0$ .

This paper is organized as follows: In Section 2, we formulate the problem and the associated variational principle that forms the basis of our analysis and numerical computations. In Section 3, we give general formulae for the average energy in an elastomer–ferromagnet composite. We also derive the  $\Phi^2$  expansion of the overall energy. In Section 4.1, we solve the problem of elastic interaction between pairs of rigid spheres embedded in the elastomer. In Section 4.2, we calculate the magnetic force of interaction between particles in the composite. In Section 4.3, we calculate the state of mechanical equilibrium in the composite. Displacements of the inclusions are calculated from force and torque balance equations and the distribution of magnetization in the composite is the minimizer of magnetic energy. In Section 5, we collect the results of previous sections and give the final formulas for the calculation of average energy and strain in the composite, correct to order  $\Phi^2$ . A variety of numerical results are given in Section 6.

## 2. Formulation of the problem

We deal with composite materials consisting of magnetically permeable inclusions firmly embedded in a non-magnetic elastic matrix. The inclusions are assumed to be rigid ferromagnetic spheres of radius  $a$  (Bonnecaze and Brady, 1992; Ginder and Davis, 1994; Jolly et al., 1996a). The matrix material is assumed to be homogeneous, isotropic and linearly elastic with shear modulus  $\mu$  and the Poisson ratio  $\nu$ .

### 2.1. The magneto-elastic equations

Let us consider a sample volume  $V$  of an elastomer–ferromagnet composite, containing a number  $N$  of ferromagnetic particles  $\Omega_p$  ( $p = 1, \dots, N$ ). The mathematical magneto-elastic problem associated with such a composite is described by

equations

$$\begin{aligned} \frac{\partial \Sigma_{ij}(\mathbf{x})}{\partial x_j} &= 0 \quad \text{for } \mathbf{x} \text{ in the elastic matrix,} \\ \mathbf{F}^{\text{el}(p)} + \mathbf{F}^{\text{mag}(p)} &= \mathbf{0}, \\ \mathcal{T}^{\text{el}(p)} + \mathcal{T}^{\text{mag}(p)} &= \mathbf{0} \quad \text{for } p = 1, \dots, N, \end{aligned} \tag{1}$$

where we have used the summation convention for repeated indices and  $\Sigma_{ij}(\mathbf{x})$  is the stress in the elastic matrix. (Note that the values of the stress in the rigid inclusions are not uniquely defined, as they depend on the Poisson ratio of the rigid phase. Of course, the inclusion stresses are not needed in our overall energy calculations.) The net elastic forces  $\mathbf{F}^{\text{el}(p)}$  and torques  $\mathcal{T}^{\text{el}(p)}$  acting on  $\Omega_p$ ,  $p = 1, \dots, N$ , are given by

$$\begin{aligned} \mathbf{F}^{\text{el}(p)} &= \mathbf{e}_i \int_{\partial\Omega_p} \Sigma_{ij}(\mathbf{x}) n_j^{(p)}(\mathbf{x}) \, ds, \\ \mathcal{T}^{\text{el}(p)} &= a \mathbf{e}_i \varepsilon_{ijk} \int_{\partial\Omega_p} n_j^{(p)}(\mathbf{x}) \Sigma_{km}(\mathbf{x}) n_m^{(p)}(\mathbf{x}) \, ds, \end{aligned} \tag{2}$$

where  $\mathbf{e}_i$ ,  $i = 1, 2, 3$ , are orthogonal, unit vectors,  $\varepsilon_{ijk}$  is the alternating tensor (Little, 1973) and  $\mathbf{n}^{(p)}$  is the outer normal to the surface  $\partial\Omega_p$ . To calculate the magnetic forces  $\mathbf{F}^{\text{mag}(p)}$  and torques  $\mathcal{T}^{\text{mag}(p)}$ , we solve the equations of magnetostatics (Jackson, 1975)

$$\begin{aligned} \nabla \times \mathbf{H}(\mathbf{x}) &= \mathbf{0}, \\ \nabla \cdot \mathbf{B}(\mathbf{x}) &= 0, \\ \mathbf{B}(\mathbf{x}) &= \mathbf{H}(\mathbf{x}) + 4\pi\mathbf{M}(\mathbf{x}), \end{aligned} \tag{3}$$

where  $\mathbf{H}(\mathbf{x})$  and  $\mathbf{B}(\mathbf{x})$  are the magnetic field and magnetic induction, respectively. The applied (constant) magnetic field  $\mathbf{H}_0$  induces a distribution of magnetization  $\mathbf{M}(\mathbf{x})$  in  $V$ . Since the matrix is not magnetic, the magnetization may be expressed in the form

$$\mathbf{M}(\mathbf{x}) = \begin{cases} \mathbf{M}^{(p)}(\mathbf{x}) & \text{if } \mathbf{x} \in \Omega_p, p = 1, \dots, N, \\ \mathbf{0} & \text{otherwise} \end{cases} \tag{4}$$

and the solution of Eq. (3) is given by

$$\mathbf{H}(\mathbf{x}) = \mathbf{H}_0 - \nabla \Psi_M(\mathbf{x}), \tag{5}$$

where  $\Psi_M$  is the scalar potential (Jackson, 1975)

$$\begin{aligned} \Psi_M(\mathbf{x}) &= \sum_{p=1}^N \Psi_M^{(p)}(\mathbf{x}), \\ \Psi_M^{(p)}(\mathbf{x}) &= \left[ - \int_{\Omega_p} \frac{\nabla' \cdot \mathbf{M}^{(p)}(\mathbf{x}')}{|\mathbf{x} - \mathbf{x}'|} \, d\mathbf{x}' + \int_{\partial\Omega_p} \frac{\mathbf{n}^{(p)}(\mathbf{x}') \cdot \mathbf{M}^{(p)}(\mathbf{x}')}{|\mathbf{x} - \mathbf{x}'|} \, ds' \right]. \end{aligned} \tag{6}$$

The magnetic force acting on  $\Omega_p$  is (Jackson, 1975)

$$\mathbf{F}^{\text{mag}(p)} = - \int_{\Omega_p} \mathbf{B}_e(\mathbf{x})[\nabla \cdot \mathbf{M}^{(p)}(\mathbf{x})] \, d\mathbf{x} + \int_{\partial\Omega_p} [\mathbf{M}^{(p)}(\mathbf{x}) \cdot \mathbf{n}^{(p)}(\mathbf{x})]\mathbf{B}_e(\mathbf{x}) \, ds, \quad (7)$$

where

$$\mathbf{B}_e(\mathbf{x}) = \mathbf{H}_0 - \sum_{k=1, k \neq p}^N \nabla \Psi_M^{(k)}(\mathbf{x}) \quad (8)$$

is the external magnetic induction (not including that of particle  $\Omega_p$  itself). Similarly, one can write the expression of torque  $\mathcal{T}^{\text{mag}(p)}$  in terms of  $\mathbf{M}^{(p)}$  and  $\mathbf{B}_e$  (Jackson, 1975).

To close system (3), we calculate  $\mathbf{M}(\mathbf{x})$ . Our approach is based on the fact that the magnetization distribution (4) in the composite minimizes the magnetic energy (Brown, 1962; Landau and Lifshitz, 1996)

$$\langle \mathcal{W}_t^{\text{mag}} \rangle = \frac{1}{V} \sum_{p=1}^N \left\{ \int_{\Omega_p} \left[ -\mathbf{M}^{(p)} \cdot \mathbf{H}^0 + \frac{1}{2} \mathbf{M}^{(p)} \cdot \nabla \Psi_M \right] \, d\mathbf{x} + \mathcal{W}_{\text{m-el}}^{(p)} + \mathcal{W}_{\text{non-u}}^{(p)} + \mathcal{W}_{\text{aniso}}^{(p)} \right\} \quad (9)$$

subject to the constraint that the particle magnetization cannot exceed the saturation value  $M^{\text{sat}}$  (Morrish, 1965)

$$|\mathbf{M}^{(p)}| \leq M^{\text{sat}} \quad \text{for } p = 1, \dots, N. \quad (10)$$

The integral in the right-hand side of Eq. (9) describes the energy of the particles' magnetization in the applied field  $\mathbf{H}_0$  and the self-energy of the magnetization in its own field, respectively; the quantity  $\mathcal{W}_{\text{m-el}}^{(p)}$ , in turn, is the magnetostriction energy which accounts for deformation of a ferromagnet due to changes in its magnetization. In general, further, a ferromagnet has a domain structure (Landau and Lifshitz, 1996; Morrish, 1965) and  $\mathcal{W}_{\text{non-u}}^{(p)}$  is the additional energy due to the non-uniformity of the magnetization. (A magnetic domain is a region of constant magnetization and a ferromagnet can contain many domains, each one with a different magnetization. Adjacent domains are separated by domain walls, which are thin layers where the magnetization changes continuously from one domain to another.) Finally,  $\mathcal{W}_{\text{aniso}}^{(p)}$  is the magnetic anisotropy energy which depends on the direction of magnetization. In uniaxial ferromagnets, such as hexagonal cobalt, the anisotropy energy is an important term that has to be taken into account. In cubic crystals, such as iron and iron–cobalt alloys, the anisotropy energy is weaker and it can be neglected for applied magnetic fields  $H^0 \sim 4\pi M^{\text{sat}}$  or higher (Landau and Lifshitz, 1996).

Typically the magneto-elastic energy  $\mathcal{W}_{\text{m-el}}^{(p)}$  is much smaller (Landau and Lifshitz, 1996) than other terms in Eq. (9) and it is therefore neglected in this paper. Further, in this study, we assume that magnetizations are constant within each one of the ferromagnetic particles:  $\mathbf{M}^{(p)}$  is a constant vector for  $p = 1, \dots, N$ . As discussed in the introduction, this assumption is justified in a number of situations, including cases

in which particles are sufficiently small (diameter  $\leq 1.5 \mu\text{m}$  for iron (Landau and Lifshitz, 1996; Morrish, 1965)), so that either magnetic domains cannot form at all, or that magnetization variations within each particle are restricted to form a structure consisting of thin layers (a laminate) which generates a magnetic field that is effectively equivalent to the one induced by a constant magnetization—with strength equal to the average value of the magnetization within the layered structure. Due to our assumption of uniformly magnetized particles, we set in Eq. (9)  $\mathcal{W}_{\text{non-}u}^{(p)} = 0$  and, after neglecting the magnetostriction and anisotropy energies, we obtain the simplified energy expression we use:

$$\langle \mathcal{W}^{\text{mag}} \rangle = \frac{1}{V} \sum_{p=1}^N \left[ - \int_{\Omega_p} \mathbf{M}^{(p)} \cdot \mathbf{H}^0 \, d\mathbf{x} + \int_{\Omega_p} \frac{1}{2} \mathbf{M}^{(p)} \cdot \nabla \Psi_M(\mathbf{x}) \, d\mathbf{x} \right]. \tag{11}$$

In view of the previous discussion, the approximations involved in this expression can generally be expected to provide very accurate results for small particles and large applied magnetic fields (particle diameters  $\leq 1.5 \mu\text{m}$  for iron and  $H^0 \sim 4\pi M^{\text{sat}}$  or higher). Further, correct order-of-magnitude estimates should result rather generally. The additional complexity required to incorporate all of the effects neglected in this energy expression requires numerical evaluation of magnetizations and interparticle forces—which, in the present simplified context, we are able to evaluate in closed form. Such complete numerical treatments, which should not require inordinately long simulations, would constitute a valuable continuation of the present work.

The displacement of particles  $\Omega_p$ ,  $p = 1, \dots, N$ , consists of rigid body translations and rotations,

$$\begin{aligned} \mathbf{u}(\mathbf{x}) &= a\mathbf{T}^{(p)} + \mathbf{R}^{(p)} \times \mathbf{r}^{(p)} \\ &= aT_j^{(p)} \mathbf{e}_j + R_j^{(p)} \varepsilon_{ijk} (\mathbf{r}^{(p)} \cdot \mathbf{e}_k) \mathbf{e}_i, \quad \text{for } \mathbf{x} \in \Omega_p, \quad p = 1, \dots, N, \end{aligned} \tag{12}$$

where  $\mathbf{r}^{(p)}$  is the vector position of  $\mathbf{x}$  with respect to  $\mathbf{x}_u^{(p)}$ , the center of  $\Omega_p$  in the undeformed configuration. Here,  $T_j^{(p)}$  quantify rigid body translations of  $\Omega_p$ , in direction  $\mathbf{e}_j$ . Furthermore,  $R_j^{(p)}$  corresponds to a rigid body rotation around the center of  $\Omega_p$ , in the plane orthogonal to  $\mathbf{e}_j$ . Outside the particles, the stress is related to the strain  $E_{ij}(\mathbf{x})$  by Hooke’s law

$$\Sigma_{ij}(\mathbf{x}) = \lambda E_{kk}(\mathbf{x}) \delta_{ij} + 2\mu E_{ij}(\mathbf{x}), \tag{13}$$

where  $\delta_{ij}$  is the Kronecker delta and  $\lambda$  is the Lamé constant of the matrix, given in terms of the shear modulus  $\mu$  and the Poisson ratio  $\nu$  as  $\lambda = 2\mu\nu/1 - 2\nu$ . Finally, the strain is given in terms of the displacement  $\mathbf{u}(\mathbf{x})$  as

$$E_{ij}(\mathbf{x}) = \frac{1}{2} \left( \frac{\partial u_i(\mathbf{x})}{\partial x_j} + \frac{\partial u_j(\mathbf{x})}{\partial x_i} \right), \quad i, j = 1, 2, 3. \tag{14}$$

In what follows, we will evaluate the overall behavior of MR composites under given applied fields. The applied magnetic field  $\mathbf{H}_0$  is simply the value of the magnetic field at infinity. At the boundary of the composite, on the other hand, we may prescribe either the displacement

$$\mathbf{u}(\mathbf{x}) = \mathbf{u}^0(\mathbf{x}) \quad \text{for } \mathbf{x} \in \partial V, \tag{15}$$

or surface tractions

$$\Sigma_{ij}(\mathbf{x})n_j(\mathbf{x}) = S_i(\mathbf{x}) \quad \text{for } \mathbf{x} \in \partial V \quad \text{and} \quad i = 1, 2, 3, \quad (16)$$

where  $\mathbf{n}$  is the outer normal to  $\partial V$ .

## 2.2. A variational principle

Our evaluation of the overall magneto-elastic properties of composites relies upon well known explicit solutions for elastic and magnetic inclusion problems. As we will see, such solutions, which are parametrized by the values of the associated elastic displacements at infinity, lend themselves rather directly for the treatment of boundary value problems of Dirichlet type—in which homogeneous displacements are prescribed at the boundary of the composite. In the present context, however, it is of substantial interest to describe the behavior of a composite under boundary conditions of the Neumann type, for which boundary tractions are prescribed instead. It is easy to appreciate the importance of solutions of such Neumann problems: for example, the evaluation of the maximum strains achievable by a composite requires solution of the magneto-elastic problem under *zero* applied tractions. The following remarks describe a variational principle which in fact yields the elastic energy minimizations associated with Neumann problems in terms of a finite-dimensional minimization of energies corresponding to certain Dirichlet problems—for which the relevant inclusion solutions are known. A simple fact lies at the basis of the connections between variational principles for Dirichlet and Neumann problems:

**Remark 1.** Owing to the assumed uniform statistical distribution of particles within the composites under consideration, the boundary values of the displacement arising in such composites from given homogeneous tractions

$$S_i = \Sigma_{ij}n_j, \quad (17)$$

are themselves homogeneous. More precisely, with probability one, the solution  $\mathbf{u}$  of the magneto-elastic problem for homogeneous tractions  $S_i$  necessarily takes, under the infinite volume (homogenization) limit, the boundary values

$$u_i|_{\partial V} = \underline{\underline{\epsilon}}_{ij}^0 x_j, \quad (18)$$

for some constant symmetric tensor  $\underline{\underline{\epsilon}}^0$ . Further, the magneto-elastic energy associated with the solution of the boundary value problem with boundary data (18) converges, in the homogenization limit, to the homogenized energy in the original traction problem.

To establish the validity of this remark we may restrict ourselves to the purely elastic case; the microscopic magnetic forces can then be incorporated easily, by using the div-curl lemma, and by linearity of the magnetoelastic equations (1) with respect to the particle boundary conditions. To deal with the purely elastic case we recall from homogenization theory (Jikov et al., 1994; Tartar, 1977) that, calling  $C_{ijkl}^*$  the effective stiffness tensor and  $\Sigma^0$  the infinite-volume average of  $\Sigma$ , under the homogenization

limit we have

$$\Sigma_{ij}^0 = C_{ijkl}^* \varepsilon_{kl}^0$$

with probability one—for some constant tensor  $\varepsilon_{kl}^0$ . By convergence of energies (compensated compactness (Jikov et al., 1994; Tartar, 1977)) the elastic energy associated with the solution of the corresponding boundary value problem with boundary conditions (18) converges to the homogenized energy for problem (17) with probability one. Further, in the limit  $V \rightarrow \infty$  a suitable rescaling of the displacement associated with the data (17) converges (with probability one, weakly in  $H^1$  and thus strongly in  $H^s$  for  $\frac{1}{2} < s < 1$  (Adams, 1975)) to the homogeneous displacement  $\varepsilon_{ij}^0 x_j$ . It follows from the trace theorem (Adams, 1975) that the boundary values of  $\mathbf{u}$  converge strongly in  $H^{s-(1/2)} \subset L^2$  to the homogeneous displacement (18).

From the principles of minimum energy for elasticity (Sokolnikoff, 1956) and magnetism (Brown, 1962) we thus obtain our governing variational principle, which we detail in the following remark.

**Remark 2.** The solution  $\mathbf{u}(\mathbf{x})$  of the equilibrium equations (1)–(16) with boundary conditions (17) equals  $\tilde{\mathbf{v}}$ , where  $(\underline{\underline{\varepsilon}}, \tilde{\mathbf{v}}, \tilde{\mathbf{M}})$  is the minimizer of the variational principle

$$\min_{\underline{\underline{\varepsilon}}^0} \min_{v_i|_{\partial V} = \varepsilon_{ij}^0 x_j} \min_{|\mathbf{M}| \leq \mathbf{M}^{\text{sat}}} \mathcal{U}(\underline{\underline{\varepsilon}}^0, \mathbf{v}, \mathbf{M}). \tag{19}$$

Here, the potential energy  $\mathcal{U}$  is given by

$$\mathcal{U}(\underline{\underline{\varepsilon}}^0, \mathbf{v}, \mathbf{M}) = \langle \mathcal{W}^{\text{el}} \rangle + \langle \mathcal{W}^{\text{mag}} \rangle - \frac{1}{V} \int_{\partial V} S_i(\mathbf{x}) v_i(\mathbf{x}) \, ds, \tag{20}$$

where  $\langle \mathcal{W}^{\text{el}} \rangle$  is the strain energy associated with displacement vector  $\mathbf{v}$  and where, for a given applied, constant field  $\mathbf{H}_0$  and a given arrangement of the inclusions  $\Omega_p$ ,  $p = 1, \dots, N$ , the magnetic energy  $\langle \mathcal{W}^{\text{mag}} \rangle$  is given by Eq. (11). The displacement  $\mathbf{v}_{\underline{\underline{\varepsilon}}^0}$ , which is defined as the partial minimizer of the energy  $\mathcal{U}$  in Eq. (19) with respect to  $\mathbf{M}$  and  $\mathbf{v}$  for given  $\underline{\underline{\varepsilon}}^0$ , plays an important role in our numerical scheme; see also remark below.

**Remark 3.** The intermediate solution  $\mathbf{v}_{\underline{\underline{\varepsilon}}^0}(\mathbf{x})$  utilized in Remark 2 is obtained as follows. For a given, arbitrary assignment of displacements  $\mathbf{T}^{(p)}$  and rotations  $\mathbf{R}^{(p)}$  of the particles  $\Omega_p$ ,  $p = 1, \dots, N$ , we define  $\mathbf{w}(\mathbf{x})$  to be the solution of equations

$$\begin{aligned} \frac{\partial \sigma_{ij}(\mathbf{x})}{\partial x_j} &= 0, \\ \sigma_{ij}(\mathbf{x}) &= \lambda \varepsilon_{kk}(\mathbf{x}) \delta_{ij} + 2\mu \varepsilon_{ij}(\mathbf{x}), \\ \varepsilon_{ij}(\mathbf{x}) &= \frac{1}{2} \left( \frac{\partial w_i(\mathbf{x})}{\partial x_j} + \frac{\partial w_j(\mathbf{x})}{\partial x_i} \right), \quad i, j = 1, 2, 3, \quad \mathbf{x} \text{ in the elastic matrix,} \end{aligned} \tag{21}$$

for which, at the surface of particle  $\Omega_p$  ( $p = 1, \dots, N$ ) we have

$$\mathbf{w}(\mathbf{x}) = a \mathbf{T}^{(p)} + \mathbf{R}^{(p)} \times \mathbf{r}^{(p)}, \quad \mathbf{r}^{(p)} = \mathbf{x} - \mathbf{x}_u^{(p)}, \quad \mathbf{x} \in \partial \Omega_p \tag{22}$$

and which, at the outer boundary satisfies the Dirichlet boundary conditions

$$w_i(\mathbf{x}) = \varepsilon_{ij}^0 x_j \quad \text{for } \mathbf{x} \in \partial V. \tag{23}$$

Then, for a trial strain  $\underline{\underline{\varepsilon}}^0$ , the displacement  $\mathbf{v}_{\underline{\underline{\varepsilon}}^0}$  is the minimizer of  $\mathcal{U}(\underline{\underline{\varepsilon}}^0, \mathbf{w}, \mathbf{M})$  over all possible choices of particle rigid displacements and rotations  $\mathbf{T}^{(p)}$  and  $\mathbf{R}^{(p)}$  and all magnetizations with magnitude  $|\mathbf{M}| \leq M^{\text{sat}}$ .

### 3. The overall energy

In this section, we derive representations of the total strain and magnetic energies of an elastomer–ferromagnet composite in terms of certain integrals over the particle bodies. These representations assume a given boundary displacement and a given applied magnetic field. As mentioned earlier the Neumann case, in which boundary tractions are prescribed instead, will be handled through reduction to the Dirichlet case discussed in this section—through a suitable application of Remark 2.

#### 3.1. The overall strain energy

Consider a displacement field  $\mathbf{u}$  which, at the boundary of  $V$  satisfies the condition

$$u_i(\mathbf{x}) = \varepsilon_{ij}^0 x_j, \quad i = 1, 2, 3, \quad \mathbf{x} \in \partial V; \tag{24}$$

clearly, the associated average strain equals  $\varepsilon_{ij}^0$

$$\varepsilon_{ij}^0 = \langle E_{ij} \rangle = \frac{1}{2} \left\langle \frac{\partial u_i(\mathbf{x})}{\partial x_j} + \frac{\partial u_j(\mathbf{x})}{\partial x_i} \right\rangle, \quad i, j = 1, 2, 3. \tag{25}$$

Here the symbol  $\langle \cdot \rangle$  denotes volume average or ensemble average over the ensemble  $\Omega$  of all possible realizations

$$\langle \cdot \rangle = \lim_{V \rightarrow \infty} \frac{1}{V} \int_V \cdot d\mathbf{x} = \int_{\Omega} \cdot P(d\omega). \tag{26}$$

(By ergodicity, which we assume throughout, volume and ensemble averages coincide with probability one (Sinay, 1977). The limit  $V \rightarrow \infty$  in Eq. (26) is the homogenization limit—under which the characteristic length of  $V$  is much larger than the radii  $a$  of the ferromagnetic particles.)

Let us denote by  $V_M = V \setminus \bigcup_{p=1}^N \Omega_p$ , the volume occupied by the elastic matrix so that, the overall strain energy is given by

$$\begin{aligned} \langle \mathcal{W}^{\text{el}} \rangle &= \lim_{V \rightarrow \infty} \frac{1}{2V} \int_{V_M} \Sigma_{ij}(\mathbf{x}) E_{ij}(\mathbf{x}) d\mathbf{x} = \lim_{V \rightarrow \infty} \frac{1}{2V} \int_{V_M} \Sigma_{ij}(\mathbf{x}) \frac{\partial u_i(\mathbf{x})}{\partial x_j} d\mathbf{x} \\ &= \lim_{V \rightarrow \infty} \frac{1}{2V} \left\{ \int_{\partial V} \Sigma_{ij}(\widehat{\mathbf{x}}) n_j(\widehat{\mathbf{x}}) u_i(\widehat{\mathbf{x}}) ds - \sum_{p=1}^N \int_{\partial \Omega_p} u_i(\mathbf{x}) \Sigma_{ij}(\mathbf{x}) n_j^{(p)}(\mathbf{x}) ds \right\}. \end{aligned} \tag{27}$$

From Eq. (24) and identity

$$\int_{\partial V} x_j \Sigma_{ip}(\mathbf{x}) n_p(\mathbf{x}) \, ds = \int_{V_M} \Sigma_{ij}(\mathbf{x}) \, d\mathbf{x} + \sum_{p=1}^N \int_{\partial \Omega_p} x_j \Sigma_{ik}(\mathbf{x}) n_k^{(p)}(\mathbf{x}) \, ds,$$

we have

$$\langle \mathcal{W}^{el} \rangle = \lim_{V \rightarrow \infty} \frac{1}{2V} \left\{ \varepsilon_{ij}^0 \int_{V_M} \Sigma_{ij}(\mathbf{x}) \, d\mathbf{x} + \sum_{p=1}^N \int_{\partial \Omega_p} [\varepsilon_{ij}^0 x_j - u_i(\mathbf{x})] \Sigma_{ik}(\mathbf{x}) n_k^{(p)}(\mathbf{x}) \, ds \right\}. \tag{28}$$

Finally, from Hooke’s law,

$$\begin{aligned} \int_{V_M} \Sigma_{ij}(\mathbf{x}) \, d\mathbf{x} &= \int_{V_M} [\lambda E_{kk}(\mathbf{x}) \delta_{ij} + 2\mu E_{ij}(\mathbf{x})] \, d\mathbf{x} \\ &= V \lambda \varepsilon_{kk}^0 \delta_{ij} + 2V \mu \varepsilon_{ij}^0 - \sum_{p=1}^N \int_{\partial \Omega_p} \{ \lambda u_k(\mathbf{x}) n_k^{(p)}(\mathbf{x}) \delta_{ij} \\ &\quad + \mu [u_i(\mathbf{x}) n_j^{(p)}(\mathbf{x}) + u_j(\mathbf{x}) n_i^{(p)}(\mathbf{x})] \} \, ds \end{aligned}$$

so the overall strain energy becomes

$$\begin{aligned} \langle \mathcal{W}^{el} \rangle &= \frac{\lambda}{2} (\varepsilon_{kk}^0)^2 + \mu \varepsilon_{ij}^0 \varepsilon_{ij}^0 + \lim_{V \rightarrow \infty} \frac{1}{2V} \sum_{p=1}^N \int_{\partial \Omega_p} \{ [\varepsilon_{ij}^0 x_j - u_i(\mathbf{x})] \Sigma_{ik}(\mathbf{x}) n_k^{(p)}(\mathbf{x}) \\ &\quad - \lambda \varepsilon_{ii}^0 u_k(\mathbf{x}) n_k^{(p)}(\mathbf{x}) - \mu \varepsilon_{ij}^0 [u_i(\mathbf{x}) n_j^{(p)}(\mathbf{x}) + u_j(\mathbf{x}) n_i^{(p)}(\mathbf{x})] \} \, ds. \end{aligned} \tag{29}$$

The overall energy depends, among other factors, on the distribution of particles in the composite; in what follows we assume a random, statistically homogeneous distribution of  $N \gg 1$  particles in the sample volume  $V$ . To obtain a convenient description of the dependence of the overall energy on the statistics of the composite we re-express Eq. (29) as a statistical average of the energy content in a “reference particle”  $\Omega_1$  (Jeffrey, 1973; Willis and Acton, 1976). In detail, by ergodicity we may write

$$\langle \mathcal{W}^{el} \rangle = \frac{1}{2} [\lambda (\varepsilon_{kk}^0)^2 + 2\mu \varepsilon_{ij}^0 \varepsilon_{ij}^0] + \mathcal{N} \overline{\mathcal{W}^{el(1)}}, \tag{30}$$

where  $\mathcal{N}$  is the particle density

$$\mathcal{N} = \frac{N}{V} = \frac{\Phi}{4\pi a^3/3} \tag{31}$$

and where  $\overline{\mathcal{W}^{el(1)}}$  is the statistical average of quantity

$$\begin{aligned} \mathcal{W}^{el(1)} &= \int_{\partial \Omega_1} \{ [\varepsilon_{ij}^0 x_j - u_i(\mathbf{x})] \Sigma_{ik}(\mathbf{x}) n_k^{(1)}(\mathbf{x}) - \lambda \varepsilon_{ii}^0 u_k(\mathbf{x}) n_k^{(1)}(\mathbf{x}) \\ &\quad - \mu \varepsilon_{ij}^0 [u_i(\mathbf{x}) n_j^{(1)}(\mathbf{x}) + u_j(\mathbf{x}) n_i^{(1)}(\mathbf{x})] \} \, ds, \end{aligned} \tag{32}$$

over all configurations containing a particle  $\Omega_1$  centered at the origin.

Using the particle centers  $\mathcal{C}_{\mathcal{N}} = \{\mathbf{x}^{(1)}, \mathbf{x}^{(2)}, \dots, \mathbf{x}^{(N)}\}$  to characterize a configuration of  $N$  particles and denoting by  $d\mathcal{C}_{\mathcal{N}}$  the volume element  $d\mathbf{x}^{(2)}, \dots, d\mathbf{x}^{(N)}$  of the last  $N - 1$  of the center coordinates, the average energy over all configurations containing  $N$  particles may be expressed in the form

$$\overline{\mathcal{W}_N^{\text{el}(1)}} = \int \mathcal{W}^{\text{el}(1)}(\mathcal{C}_N) P(\mathcal{C}_N|0) d\mathcal{C}_N, \tag{33}$$

where  $P(\mathcal{C}_N|0)$  is the probability density for the configuration  $\mathcal{C}_N$  given that particle  $\Omega_1$  lies at the origin:

$$\int P(\mathcal{C}_N|0) d\mathcal{C}_N = 1. \tag{34}$$

Naturally, the average value  $\overline{\mathcal{W}_N^{\text{el}(1)}}$  is obtained as a further weighted average of the quantity  $\mathcal{W}_N^{\text{el}(1)}$ . We note in passing that, in the limit in which the last  $N - 1$  particles in  $\mathcal{C}_N$  are far away from  $\Omega_1$ , the randomness and the lack of long-range order in the composite imply

$$P(\mathcal{C}_N|0) = P(\mathcal{C}_N), \tag{35}$$

where  $P(\mathcal{C}_N)$  is the probability density of the configuration  $\mathcal{C}_N$  with  $\Omega_1$  removed from the system.

### 3.2. The overall magnetic energy

We now derive expressions for the magnetostatic energy (11) stored in an array of uniformly magnetized spherical particles. To do this, we use the magnetic potential  $\Psi_M(\mathbf{x}) = \sum_{p=1}^N \Psi_M^{(p)}(\mathbf{x})$  which, for constant magnetizations  $\mathbf{M}^{(p)}$  and spherical particles  $\Omega_p$  centered at  $\mathbf{x}^{(p)}$ , is given by (see Eq. (6) and Jackson (1975))

$$\Psi_M^{(p)}(\mathbf{x}) = \begin{cases} \frac{4\pi}{3} \left( \frac{a}{|\mathbf{x} - \mathbf{x}^{(p)}|} \right)^3 \mathbf{M}^{(p)} \cdot (\mathbf{x} - \mathbf{x}^{(p)}) & \text{if } |\mathbf{x} - \mathbf{x}^{(p)}| > a, \\ \frac{4\pi}{3} \mathbf{M}^{(p)} \cdot (\mathbf{x} - \mathbf{x}^{(p)}) & \text{if } |\mathbf{x} - \mathbf{x}^{(p)}| \leq a, \quad p = 1, \dots, N. \end{cases} \tag{36}$$

The corresponding value of the magnetic field is

$$\mathbf{H}(\mathbf{x}) = \mathbf{H}_0 + \sum_{p=1}^N \mathbf{H}^{(p)}(\mathbf{x}), \tag{37}$$

where  $\mathbf{H}^{(p)}$  is given by

$$\mathbf{H}^{(p)}(\mathbf{x}) = \begin{cases} -\frac{4\pi}{3} \mathbf{M}^{(p)} & \text{if } |\mathbf{x} - \mathbf{x}^{(p)}| \leq a, \\ \frac{4\pi}{3} \left( \frac{a}{|\mathbf{x} - \mathbf{x}^{(p)}|} \right)^3 \left\{ \frac{3[\mathbf{M}^{(p)} \cdot (\mathbf{x} - \mathbf{x}^{(p)})]}{|\mathbf{x} - \mathbf{x}^{(p)}|^2} (\mathbf{x} - \mathbf{x}^{(p)}) - \mathbf{M}^{(p)} \right\} & \text{if } |\mathbf{x} - \mathbf{x}^{(p)}| > a; \end{cases} \tag{38}$$

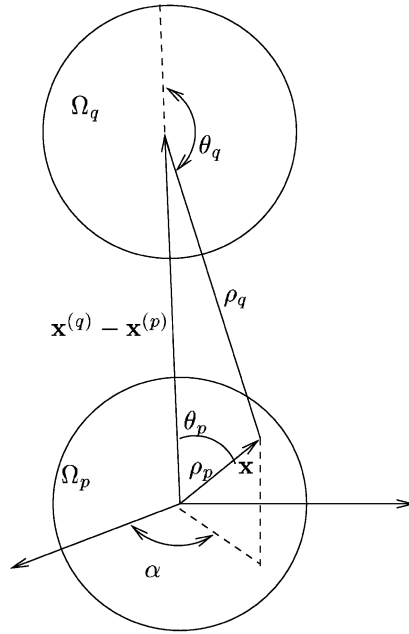


Fig. 1. Spherical systems of coordinates centered at  $\mathbf{x}^{(p)}$  and  $\mathbf{x}^{(q)}$ , respectively.

the magnetic energy, in turn, equals

$$\langle \mathcal{W}^{\text{mag}} \rangle = \frac{1}{V} \left[ -\frac{4\pi}{3} a^3 \sum_{p=1}^N \mathbf{M}^{(p)} \cdot \mathbf{H}_0 + \frac{8\pi^2}{9} a^3 \sum_{p=1}^N |\mathbf{M}^{(p)}|^2 + \frac{1}{2} \sum_{p=1}^N \sum_{q \neq p} \int_{\Omega_p} \mathbf{M}^{(p)} \cdot \nabla \Psi_M^{(q)} d\mathbf{x} \right]. \tag{39}$$

In order to evaluate the integral in Eq. (39) we use spherical coordinates  $(\rho_p, \theta_p, \alpha)$  and  $(\rho_q, \theta_q, \alpha)$  centered at  $\mathbf{x}^{(p)}$  and  $\mathbf{x}^{(q)}$ , respectively; see Fig. 1. Further, we call  $\gamma_q$  the angle between  $\mathbf{x} - \mathbf{x}^{(q)}$  and  $\mathbf{M}^{(q)}$ —so that, for  $\mathbf{x} \in \Omega_p$ , the potential  $\Psi_M^{(q)}(\mathbf{x})$  is given by

$$\Psi_M^{(q)}(\mathbf{x}) = \frac{4\pi}{3} \frac{a^3}{|\mathbf{x} - \mathbf{x}^{(q)}|^2} |\mathbf{M}^{(q)}| \cos \gamma_q. \tag{40}$$

Finally, we denote by  $\theta^{(q)} \in [0, \pi]$  the angle between  $\mathbf{M}^{(q)}$  and  $\mathbf{x}^{(q)} - \mathbf{x}^{(p)}$ , and we let  $\alpha^{(q)} \in [0, 2\pi]$  be the angle swept by the projection of  $\mathbf{M}^{(q)}$  on the plane orthogonal to  $\mathbf{x}^{(q)} - \mathbf{x}^{(p)}$ —measured with respect to an arbitrary axis orthogonal to  $\mathbf{x}^{(q)} - \mathbf{x}^{(p)}$ . With these notations, the magnetic potential (40) can be expressed as (Jackson, 1975)

$$\Psi_M^{(q)}(\mathbf{x}) = \frac{4\pi}{3} |\mathbf{M}^{(q)}| \frac{a^3}{\rho_q^2} \sum_{m=-1}^1 \frac{(1-m)!}{(1+m)!} P_1^m(\cos \theta^{(q)}) P_1^m(\cos \theta_q) e^{-im(\alpha - \alpha^{(q)})}, \tag{41}$$

where  $P_l^m(\cdot)$  are the Associated Legendre functions (Abramovitz and Stegun, 1972) ( $l = 1$  in Eq. (41)). Using the addition theorem (Hobson, 1931)

$$\frac{P_1^m(\cos \theta_q)}{\rho_q^2} = (-1)^{m+1} \sum_{s=m}^{\infty} \frac{(s+1)!}{(s+m)!(1-m)!} \frac{\rho_p^s}{|\mathbf{x}^{(q)} - \mathbf{x}^{(p)}|^{s+2}} P_s^m(\cos \theta_p) \quad (42)$$

we obtain

$$\Psi_M^{(q)}(\mathbf{x}) = \frac{4\pi}{3} |\mathbf{M}^{(q)}| a^3 \left[ P_1^1(\cos \theta^{(q)}) \cos(\alpha - \alpha^{(q)}) \sum_{s=1}^{\infty} \frac{\rho_p^s}{|\mathbf{x}^{(q)} - \mathbf{x}^{(p)}|^{s+2}} P_s^1(\cos \theta_p) - P_1(\cos \theta^{(q)}) \sum_{s=0}^{\infty} \frac{(s+1)\rho_p^s}{|\mathbf{x}^{(q)} - \mathbf{x}^{(p)}|^{s+2}} P_s(\cos \theta_p) \right]. \quad (43)$$

The total energy is now evaluated directly:

$$\langle \mathcal{W}^{\text{mag}} \rangle = \frac{1}{V} \sum_{p=1}^N \mathcal{W}^{\text{mag}(p)}, \quad (44)$$

where

$$\begin{aligned} \mathcal{W}^{\text{mag}(p)} = & -\frac{4\pi}{3} a^3 \mathbf{M}^{(p)} \cdot \mathbf{H}_0 + \frac{8\pi^2}{9} a^3 |\mathbf{M}^{(p)}|^2 + \frac{8\pi^2}{9} a^3 \sum_{q \neq p} \left( \frac{a}{|\mathbf{x}^{(q)} - \mathbf{x}^{(p)}|} \right)^3 \\ & \times \left[ \mathbf{M}^{(p)} \cdot \mathbf{M}^{(q)} - 3 \frac{\mathbf{M}^{(p)} \cdot (\mathbf{x}^{(q)} - \mathbf{x}^{(p)})}{|\mathbf{x}^{(q)} - \mathbf{x}^{(p)}|} \frac{\mathbf{M}^{(q)} \cdot (\mathbf{x}^{(q)} - \mathbf{x}^{(p)})}{|\mathbf{x}^{(q)} - \mathbf{x}^{(p)}|} \right]. \quad (45) \end{aligned}$$

### 3.3. Strain energy: second order expansion

To evaluate the average strain energy  $\langle \mathcal{W}^{\text{el}} \rangle$  of Eq. (30) up to second order in powers of the volume fraction  $\Phi$ , we note that only particles that lie within a distance of  $O(a)$  from  $\Omega_1$  produce a  $O(\Phi)$  effect in  $\mathcal{W}^{\text{el}(1)}$  (Batchelor and Green, 1972; Chen and Acrivos, 1978b; Jeffrey, 1974; Willis and Acton, 1976) Indeed, the probability that one particle is within a distance  $O(a)$  from  $\Omega_1$  is  $O(\Phi)$ , whereas the probability that two or more particles are at distance  $O(a)$  from  $\Omega_1$  is of order  $\Phi^2$ . In view of Eq. (30), then, an evaluation of the second order expansion of  $\langle \mathcal{W}^{\text{el}} \rangle$  need only consider the former particle–particle interaction case. That is, the displacements, strains and stresses in Eq. (32) may be substituted by those arising in  $\Omega_1$  as two particles,  $\Omega_1$  and, say,  $\Omega_2$ , lie within the infinite elastic matrix. Using such a pair  $\{\Omega_1, \Omega_2\}$  of particles, (with  $\Omega_2$  located at point  $\mathbf{r}$  measured with respect to  $\Omega_1$ ), the strain energy  $\mathcal{W}^{\text{el}(1)}$  is given by

$$\mathcal{W}^{\text{el}(1)}(\mathcal{C}) \approx \mathcal{W}_{1-2}^{\text{el}(1)}(\mathbf{r}), \quad (46)$$

where  $\mathcal{W}_{1-2}^{\text{el}(1)}$  denotes the value expression (32) takes when the displacements, strains and stresses in its integrand are substituted by those arising in the two-particle system. Performing the integration (30) we then obtain

$$\langle \mathcal{W}^{\text{el}} \rangle = \frac{1}{2} [\lambda (\varepsilon_{kk}^0)^2 + 2\mu \varepsilon_{ij}^0 \varepsilon_{ij}^0] + \mathcal{N} \int \mathcal{W}_{1-2}^{\text{el}(1)}(\mathbf{r}) P(\mathbf{r}|\mathbf{0}) \, d\mathbf{r} + o(\Phi^2), \quad (47)$$

where  $P(\mathbf{r}|0)$  denotes the probability density of finding a particle at  $\mathbf{r}$  given that there is a particle centered at the origin.

We note that all the statistical information necessary to evaluate the overall strain energy to second order in the volume fraction is encoded in the probability density  $P(\mathbf{r}|0)$ . For a statistically homogeneous composite, the probability that a particle can be found at location  $\mathbf{r}$  is

$$P(\mathbf{r}) = \mathcal{N}. \quad (48)$$

The probability density of finding particle  $\Omega_2$  at  $\mathbf{r}$  when  $\Omega_1$  is at the origin,  $P(\mathbf{r}|0)$ , depends on the statistical character of the composite microstructure. We adopt the hypothesis of a well-stirred suspension,

$$P(\mathbf{r}|0) = \begin{cases} 0 & \text{if } |\mathbf{r}| < 2a, \\ \mathcal{N} & \text{if } |\mathbf{r}| \geq 2a, \end{cases} \quad (49)$$

where  $\Omega_2$  can be found with equal probability in all accessible positions (Batchelor, 1972; Jeffrey, 1973). Other probability densities  $P(\mathbf{r}|0)$  of anisotropic distributions can be considered as well, without any change to the method of analysis presented in this paper.

We end this section with a note about the integral in Eq. (47), which, as it stands, is not absolutely convergent. To obtain a convergent integral, we correct Eq. (47) as in Batchelor and Green (1972), Chen and Acrivos (1978b), Hinch (1977), Jeffrey (1973, 1974). Explicitly, we show in Section 5.1 (see Eqs. (93)–(95) and (97)) that the average strain energy  $\langle \mathcal{W}^{\text{el}} \rangle$  can be written as a sum of two terms: The first term is due to the magnetic interactions in  $V$  (it vanishes if  $\mathbf{H}_0 = 0$ ) and it is absolutely convergent. The second term is  $\lambda^*/2(\epsilon_{ii}^0)^2 + \mu^* \epsilon_{ij}^0 \epsilon_{ij}^0$ , where  $\lambda^*$  and  $\mu^*$  are the effective Lamé constant and shear modulus of the composite, in the absence of an applied magnetic field. The difficulty posed by the conditional convergence of the integral in Eq. (47) is encountered in the calculation to  $O(\Phi^2)$  of the effective parameters  $\lambda^*$  and  $\mu^*$  or, equivalently, of the average stress  $\langle \tilde{\sigma}_{ij} \rangle = \lambda^* \epsilon_{kk}^0 \delta_{ij} + 2\mu^* \epsilon_{ij}^0$ . We take  $\lambda^*$  and  $\mu^*$ , correct to  $O(\Phi^2)$ , as given in Chen and Acrivos (1978), where the authors make the corrections needed to eliminate the conditional convergence in Eq. (47).

### 3.4. Magnetic energy: second order expansion

We now evaluate the second order expansion of the average magnetic energy obtained in Section 3.2. To do this we rewrite Eq. (44) in the form

$$\langle \mathcal{W}^{\text{mag}} \rangle = \frac{\mathcal{N}}{N} \sum_{p=1}^N \mathcal{W}^{\text{mag}(p)} = \mathcal{N} \overline{\mathcal{W}^{\text{mag}(1)}}, \quad (50)$$

where  $\Omega_1$  is the reference particle which, according to our conventions, is centered at  $\mathbf{x}^{(1)} = 0$ . As pointed out in Section 3.3, the probability that one particle is within a distance of order  $a$  from  $\Omega_1$  is of order  $\Phi$ , and the probability that two or more particles are within a distance of order  $a$  from  $\Omega_1$  is of order  $\Phi^2$ . As we will show, to produce an approximation of  $\langle \mathcal{W}^{\text{mag}} \rangle$  up to order  $\Phi^2$  we may use the approximate

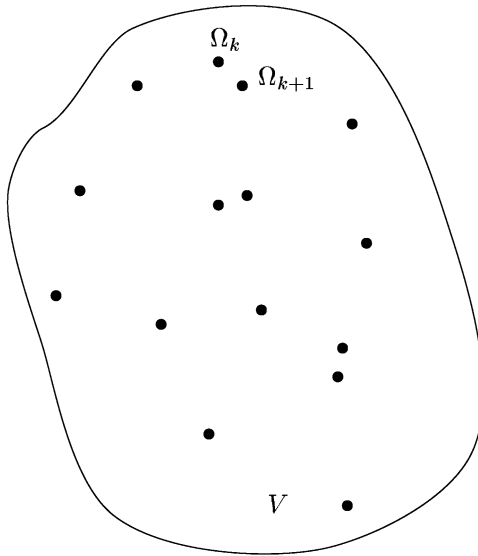


Fig. 2. Typical configuration of a dilute suspension of particles in a volume  $V$ .

value of  $\mathcal{H}^{\text{mag}(p)}$  which results as we neglect in Eq. (45) the contributions from all particles except the one nearest to  $\Omega_p$  (see Fig. 2). Note that this conclusion requires proof, since the double sum arising from Eqs. (44) and (45) is not convergent.

To show that neglecting all but the nearest particles does indeed lead to an approximation of second order in the volume fraction, let us consider a configuration of particles  $\Omega_1, \Omega_2, \dots, \Omega_N$ . The particle  $\Omega_1$  is within a distance of order  $a$  from its closest neighbor, say  $\Omega_2$ ; the remaining particles,  $\Omega_q, q \geq 3$ , are located at distances  $|\mathbf{x}^{(1)} - \mathbf{x}^{(q)}| \geq a/\delta$ , where  $\delta \ll 1$ . The magnetic force acting on particle  $\Omega_1$  is given by

$$\mathbf{F}^{\text{mag}(1)} = \sum_{q=2}^N \int_{\partial\Omega_1} [\mathbf{M}^{(1)} \cdot \mathbf{n}^{(1)}(\mathbf{x})] \mathbf{H}^{(q)}(\mathbf{x}) \, ds = \sum_{q=2}^N \mathbf{f}_{1-q}(\mathbf{x}^{(1)} - \mathbf{x}^{(q)}), \tag{51}$$

where the magnetic field  $\mathbf{H}^{(q)}$ , created by particle  $\Omega_q$ , is defined by Eq. (38). In Section 4.2, we obtain

$$\mathbf{f}_{1-q}(\mathbf{x}^{(1)} - \mathbf{x}^{(q)}) = -\frac{16\pi^2 a^2}{3} \left( \frac{a}{|\mathbf{x}^{(q)} - \mathbf{x}^{(1)}|} \right)^4 \mathbf{M}^{(1)} \cdot \mathcal{A} \mathbf{M}^{(q)}, \tag{52}$$

where  $\mathcal{A}$  is a  $3 \times 3$  matrix of order one. We have

$$\sum_{q=3}^N \mathbf{f}_{1-q} = -\frac{16\pi^2 a^2}{3} \mathcal{N} \int_{|\mathbf{x} - \mathbf{x}^{(1)}| \geq \frac{a}{\delta}} \left( \frac{a}{|\mathbf{x} - \mathbf{x}^{(1)}|} \right)^4 \mathbf{M}^{(1)} \cdot \mathcal{A} \mathbf{M}(\mathbf{x}) \, d\mathbf{x} = O(\Phi\delta) \tag{53}$$

and so,

$$\mathbf{F}^{\text{mag}(1)} = \mathbf{f}_{1-2}(\mathbf{x}^{(1)} - \mathbf{x}^{(2)}) + o(\Phi). \tag{54}$$

Similarly, the force acting on particle  $\Omega_2$  is

$$\mathbf{F}^{\text{mag}(2)} = \mathbf{f}_{2-1}(\mathbf{x}^{(1)} - \mathbf{x}^{(2)}) + o(\Phi) = -\mathbf{f}_{1-2}(\mathbf{x}^{(1)} - \mathbf{x}^{(2)}) + o(\Phi). \tag{55}$$

The magnetizations  $\mathbf{M}^{(p)}$ ,  $p = 1, \dots, N$  minimize the magnetic energy (50) for the given spatial distribution of ferromagnetic particles. Suppose that the system is in magneto-elastic equilibrium, in such a way that the elastic and magnetic forces on  $\Omega_1$  and  $\Omega_2$  are in balance. As shown by Eqs. (54) and (55) and Sections 4.1 and 4.2, the forces  $\delta\mathbf{F}^{(1)}$  and  $\delta\mathbf{F}^{(2)}$  exerted on  $\Omega_1$  and  $\Omega_2$ , respectively, by magnetic and elastic interactions with  $\Omega_q$ ,  $q \geq 3$ , are of order  $o(\Phi)$ . Thus, if all particles  $\Omega_q$ ,  $q \geq 3$  are removed from the system and artificial forces  $\delta\mathbf{F}^{(1)}$  and  $\delta\mathbf{F}^{(2)}$  are made to act on  $\Omega_1$  and  $\Omega_2$ , the system consisting of these two particles in the overall elastic matrix remains in magneto-elastic equilibrium—with unchanged values of the magnetizations  $\mathbf{M}^{(1)}$  and  $\mathbf{M}^{(2)}$ . It follows that the energy associated with this reduced system

$$\mathcal{W}_{1-2} + \int_{\mathbf{x}^{(1)}}^{\mathbf{x}} \delta\mathbf{F}^{(1)}(\mathbf{y}) \cdot d\mathbf{y} + \int_{\mathbf{x}^{(2)}}^{\mathbf{x}} \delta\mathbf{F}^{(2)}(\mathbf{y}) \cdot d\mathbf{y} \tag{56}$$

is minimized. Here,  $\mathcal{W}_{1-2}$  is the strain and magnetic energy of the system with particles  $\Omega_1$  and  $\Omega_2$  in an infinite matrix and  $\mathbf{x}$  is a point of reference. Therefore, magnetizations  $\mathbf{M}^{(1)}$  and  $\mathbf{M}^{(2)}$  are given by the magnetization of just two particles  $\Omega_1$  and  $\Omega_2$  in an infinite matrix plus an  $o(\Phi)$  correction.

Denoting by  $\Omega_{q(p)}$  the particle which is closest to  $\Omega_p$ , we now introduce an energy expression associated with the pair  $(\Omega_p, \Omega_{q(p)})$ :

$$\begin{aligned} \mathcal{V}^{\text{mag}(p)} = & -\frac{4\pi}{3}a^3\mathbf{M}^{(p)} \cdot \mathbf{H}_0 + \frac{8\pi^2}{9}a^3|\mathbf{M}^{(p)}|^2 + \frac{8\pi^2}{9}a^3 \left( \frac{a}{|\mathbf{x}^{(q(p))} - \mathbf{x}^{(p)}|} \right)^3 \\ & \times \left[ \mathbf{M}^{(p)} \cdot \mathbf{M}^{(q(p))} - 3 \frac{\mathbf{M}^{(p)} \cdot (\mathbf{x}^{(q(p))} - \mathbf{x}^{(p)})}{|\mathbf{x}^{(q(p))} - \mathbf{x}^{(p)}|} \frac{\mathbf{M}^{(q(p))} \cdot (\mathbf{x}^{(q(p))} - \mathbf{x}^{(p)})}{|\mathbf{x}^{(q(p))} - \mathbf{x}^{(p)}|} \right]. \end{aligned} \tag{57}$$

(Note that, with probability one, there will be only one particle at minimum distance from a given particle  $\Omega_p$ .) We have thus shown that, up to an arbitrary constant, expression (50) is approximated to order  $\Phi^2$  by the corresponding sum of pair energies

$$\frac{1}{V} \sum_{p=1}^N \mathcal{V}^{\text{mag}(p)}. \tag{58}$$

In particular, the observable strains, stresses, magnetizations arising from the substitution of Eq. (50) by Eq. (58) are correct up to and including the order  $\Phi^2$ .

As a final comment, we point out that when all particles are sufficiently far from  $\Omega_p$  we have the approximation

$$\mathcal{V}^{\text{mag}(p)} = -\frac{4\pi}{3}a^3\mathbf{M}^{(p)} \cdot \mathbf{H}_0 + \frac{8\pi^2}{9}a^3|\mathbf{M}^{(p)}|^2. \tag{59}$$

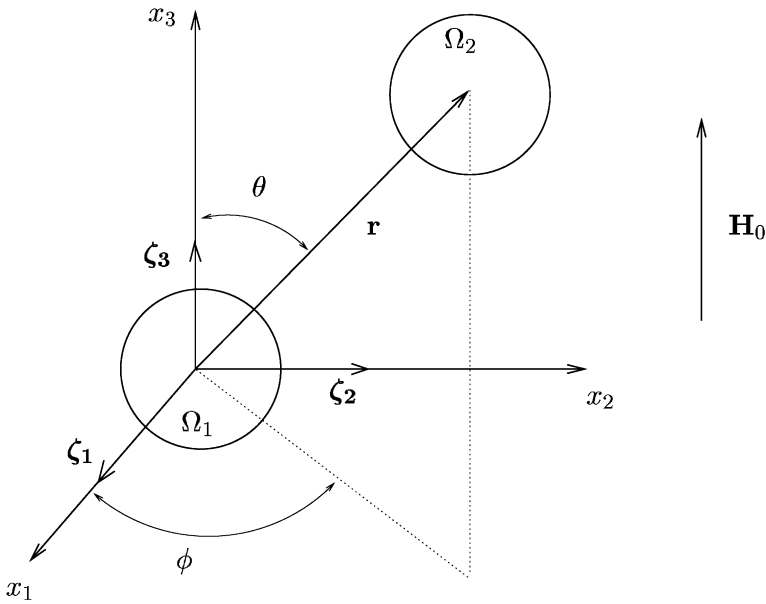


Fig. 3. Two particles, a distance  $r$  apart, embedded in an infinite matrix. The fixed system of coordinates  $(x_1, x_2, x_3)$  with unit vectors  $\zeta_i, i = 1, 2, 3$ , has the origin at the center of the reference sphere  $\Omega_1$ . The applied magnetic field is  $\mathbf{H}_0 = H_0\zeta_3$ .

In this case, Eq. (54) gives  $\mathbf{F}^{\text{mag}(p)} = o(\Phi)$  and the magnetization  $\mathbf{M}^{(p)}$ , that minimizes Eq. (59), is  $\mathbf{M}^{(p)} = \mathbf{M}^*$ , where

$$\mathbf{M}^* = \begin{cases} \frac{3}{4\pi}\mathbf{H}_0 & \text{below saturation,} \\ M^{\text{sat}} \frac{\mathbf{H}_0}{|\mathbf{H}_0|} & \text{at saturation.} \end{cases} \tag{60}$$

#### 4. Magneto-elastic fields and forces for particle pairs

As shown in Section 3, the calculation of average energy requires knowledge of the displacement  $\mathbf{u}$ , stress  $\Sigma_{ij}$  and magnetic force in systems consisting of pairs of rigid particles within an elastic matrix. We calculate these quantities by taking into account the coupled elastic and magnetic interaction between two particles  $\Omega_1$  and  $\Omega_2$ , firmly embedded in an infinite matrix.

##### 4.1. Elastic interaction

Consider the pair of particles shown in Fig. 3. The system of coordinates  $(x_1, x_2, x_3)$ , with unit vectors  $\zeta_i, i = 1, 2, 3$  and origin at the center of the reference particle  $\Omega_1$ , is chosen so that the applied magnetic field lies along the  $Ox_3$  axis:  $\mathbf{H}_0 = H_0\zeta_3$ . It is convenient to define an additional system of coordinates  $(x, y, z)$ , with unit vectors

$\mathbf{e}_i$ ,  $i = 1, 2, 3$ , where  $\mathbf{e}_3$  is along the axis of the centers of the particles in the unperturbed state, pointing from  $\Omega_1$  towards  $\Omega_2$ . Here, the unperturbed state is defined as that occurring under zero applied elastic and magnetic fields. The unit vectors  $\mathbf{e}_i$ ,  $i = 1, 2, 3$ , are obtained by rotating  $(\zeta_1, \zeta_2, \zeta_3)$  by angles  $\theta$  and  $\phi$ , as shown in Fig. 3.

In what follows, we compute the displacement  $\mathbf{u}(\mathbf{x})$  which occurs in the elastic-matrix/two-particle system as a result of prescribed displacements at infinity *and prescribed rigid motions of the particles*. (Such prescribed body motions will be eventually chosen as appropriate energy minimizers.) In detail, we seek to obtain the solution  $\mathbf{u}(\mathbf{x})$  of the system of equations

$$\begin{aligned} \nabla[\nabla \cdot \mathbf{u}(\mathbf{x})] + (1 - 2\nu)\nabla^2\mathbf{u}(\mathbf{x}) &= 0 \quad \text{outside } \Omega_1 \text{ and } \Omega_2, \\ \mathbf{u}(\mathbf{x}) &= a[T_j^{(1)}\mathbf{e}_j + R_j^{(1)}\varepsilon_{ijk}(\mathbf{n}^{(1)} \cdot \mathbf{e}_k)\mathbf{e}_i] \quad \text{at } \partial\Omega_1, \\ \mathbf{u}(\mathbf{x}) &= a[T_j^{(2)}\mathbf{e}_j + R_j^{(2)}\varepsilon_{ijk}(\mathbf{n}^{(2)} \cdot \mathbf{e}_k)\mathbf{e}_i] \quad \text{at } \partial\Omega_2, \\ \mathbf{u}(\mathbf{x}) &\rightarrow \mathbf{u}^0 \quad \text{as } |\mathbf{x}| \rightarrow \infty, \end{aligned} \tag{61}$$

where  $\mathbf{u}^0$  is the homogeneous displacement  $u_i^0 = \varepsilon_{ij}^0 x_j$ ,  $i = 1, 2, 3$ ,  $\mathbf{n}^{(p)}$  denotes the outer normal to the boundary of  $\partial\Omega_p$ , and, where  $R_j^{(i)}$  and  $T_j^{(i)}$  quantify the prescribed rigid body rotations and displacements measured with respect to the undeformed configuration.

Chen and Acrivos (1978a), as well as other authors (Miyamoto, 1958; Sternberg and Sadowsky, 1952; Tsuchida et al., 1976) obtained solutions of the equations of linear elasticity which, written in terms of the Poisson ratio  $\nu = \lambda/2(\lambda + \mu)$ , are

$$\begin{aligned} \nabla[\nabla \cdot \tilde{\mathbf{v}}(\mathbf{x})] + [1 - 2\nu]\nabla^2\tilde{\mathbf{v}}(\mathbf{x}) &= 0 \quad \text{in the elastic matrix,} \\ \tilde{\mathbf{v}}(\mathbf{x}) &\rightarrow \mathbf{u}^0 \quad \text{as } |\mathbf{x}| \rightarrow \infty. \end{aligned} \tag{62}$$

The infinite elastic matrix contains rigid particles  $\Omega_1$  and  $\Omega_2$  and the stress  $\tilde{\sigma}_{ij}$  calculated from displacement  $\tilde{\mathbf{v}}$  satisfies the force and torque balance equations

$$\begin{aligned} \mathbf{e}_i \int_{\partial\Omega_p} \tilde{\sigma}_{ij}(\mathbf{x}) n_j^{(p)}(\mathbf{x}) ds &= 0, \\ \mathbf{e}_i \varepsilon_{ijk} \int_{\partial\Omega_p} n_j^{(p)}(\mathbf{x}) \tilde{\sigma}_{km}(\mathbf{x}) n_m^{(p)}(\mathbf{x}) ds &= 0 \quad \text{for } p = 1, 2. \end{aligned} \tag{63}$$

To use these solutions as a building block in ours, we calculate the rigid body displacements induced by function  $\tilde{\mathbf{v}}$  on particles  $\Omega_1$  and  $\Omega_2$ , for a given  $\mathbf{u}^0$  at infinity:

$$\begin{aligned} \tilde{\mathbf{v}}(\mathbf{x}) &= a[\tilde{t}_j^{(1)}\mathbf{e}_j + \tilde{\omega}_j^{(1)}\varepsilon_{ijk}(\mathbf{n}^{(1)} \cdot \mathbf{e}_k)\mathbf{e}_i] \quad \text{at } \partial\Omega_1, \\ \tilde{\mathbf{v}}(\mathbf{x}) &= a[\tilde{t}_j^{(2)}\mathbf{e}_j + \tilde{\omega}_j^{(2)}\varepsilon_{ijk}(\mathbf{n}^{(2)} \cdot \mathbf{e}_k)\mathbf{e}_i] \quad \text{at } \partial\Omega_2. \end{aligned} \tag{64}$$

By linearity, the solution of Eq. (61) results as the superposition

$$\mathbf{u}(\mathbf{x}) = \tilde{\mathbf{v}}(\mathbf{x}) + \mathbf{v}(\mathbf{x}), \tag{65}$$

where  $\mathbf{v}$  satisfies the equations

$$\begin{aligned} \nabla[\nabla \cdot \mathbf{v}(\mathbf{x})] + (1 - 2\nu)\nabla^2\mathbf{v}(\mathbf{x}) &= 0 \quad \text{outside } \Omega_1 \text{ and } \Omega_2, \\ \mathbf{v}(\mathbf{x}) &= a[t_j^{(p)}\mathbf{e}_j + \omega_j^{(p)}\varepsilon_{ijk}(\mathbf{n}^{(p)} \cdot \mathbf{e}_k)\mathbf{e}_i] \quad \text{at } \partial\Omega_p, \quad p = 1, 2 \\ \mathbf{v}(\mathbf{x}) &\rightarrow \mathbf{0} \quad \text{as } |\mathbf{x}| \rightarrow \infty, \\ t_j^{(i)} &= T_j^{(i)} - \tilde{t}_j^{(i)}, \\ \omega_j^{(i)} &= R_j^{(i)} - \tilde{\omega}_j^{(i)} \quad \text{for } i = 1, 2, \quad j = 1, 2, 3. \end{aligned} \tag{66}$$

The solution of Eq. (66) can be expressed in the form

$$2\mu\mathbf{v}(\mathbf{x}) = \nabla[\tau(\mathbf{x}) + x\tau_1(\mathbf{x}) + y\tau_2(\mathbf{x}) + z\tau_3(\mathbf{x})] - 4(1 - \nu)(\tau_1(\mathbf{x}), \tau_2(\mathbf{x}), \tau_3(\mathbf{x})), \tag{67}$$

where  $\tau$  and  $\tau_i, i = 1, 2, 3$ , are the Boussinesq–Papkovitch stress functions (Eubanks and Sternberg, 1956; Naghdi and Hsu, 1961). Only three out of these four harmonic functions are independent; choices of an independent set are thus made to obtain as simple a treatment of boundary conditions as possible. We make use, once again, of the linearity of the problem and write the solution of Eq. (66) as the superposition of 12 elementary displacements

$$\begin{aligned} \mathbf{v}(\mathbf{x}) &= (t_j^{(2)} - t_j^{(1)})\boldsymbol{\chi}_j(\mathbf{x}) + (t_j^{(2)} + t_j^{(1)})\boldsymbol{\eta}_j(\mathbf{x}) + (\omega_j^{(2)} - \omega_j^{(1)})\boldsymbol{\chi}_{j+3}(\mathbf{x}) \\ &\quad + (\omega_j^{(2)} + \omega_j^{(1)})\boldsymbol{\eta}_{j+3}(\mathbf{x}), \end{aligned} \tag{68}$$

where  $\boldsymbol{\chi}_j$  and  $\boldsymbol{\eta}_j, j = 1, \dots, 6$  satisfy the differential equation defining  $\mathbf{v}$  together with conditions

$$\begin{aligned} \boldsymbol{\chi}_j &\rightarrow \mathbf{0} \quad \text{and} \quad \boldsymbol{\eta}_j \rightarrow \mathbf{0} \quad \text{at infinity} \\ \boldsymbol{\chi}_j(\mathbf{x})|_{\partial\Omega_1} &= -\frac{a}{2}\mathbf{e}_j, \quad \boldsymbol{\chi}_j(\mathbf{x})|_{\partial\Omega_2} = \frac{a}{2}\mathbf{e}_j, \\ \boldsymbol{\chi}_{j+3}(\mathbf{x})|_{\partial\Omega_1} &= -\frac{a}{2}\varepsilon_{ijk}(\mathbf{n}^{(1)} \cdot \mathbf{e}_k)\mathbf{e}_i, \quad \boldsymbol{\chi}_{j+3}(\mathbf{x})|_{\partial\Omega_2} = \frac{a}{2}\varepsilon_{ijk}(\mathbf{n}^{(2)} \cdot \mathbf{e}_k)\mathbf{e}_i, \\ \boldsymbol{\eta}_j(\mathbf{x})|_{\partial\Omega_1} &= \frac{a}{2}\mathbf{e}_j, \quad \boldsymbol{\eta}_j(\mathbf{x})|_{\partial\Omega_2} = \frac{a}{2}\mathbf{e}_j, \\ \boldsymbol{\eta}_{j+3}(\mathbf{x})|_{\partial\Omega_1} &= \frac{a}{2}\varepsilon_{ijk}(\mathbf{n}^{(1)} \cdot \mathbf{e}_k)\mathbf{e}_i, \\ \boldsymbol{\eta}_{j+3}(\mathbf{x})|_{\partial\Omega_2} &= \frac{a}{2}\varepsilon_{ijk}(\mathbf{n}^{(2)} \cdot \mathbf{e}_k)\mathbf{e}_i \quad \text{for } j = 1, 2, 3. \end{aligned} \tag{69}$$

The solution of each one of the elementary problems in Eq. (69) can be expressed as series of spherical harmonics as explained in Appendix A; the displacement  $\mathbf{v}(\mathbf{x})$  then results from Eq. (68).

The elastic force acting on particle  $\Omega_p$  is given by

$$\begin{aligned}
 F_i^{\text{el}(p)} &= \int_{\partial\Omega_p} [\sigma_{ij}(\mathbf{x}) + \tilde{\sigma}_{ij}(\mathbf{x})]n_j^{(p)}(\mathbf{x}) \, ds \\
 &= \int_{\partial\Omega_p} \sigma_{ij}(\mathbf{x})n_j^{(p)}(\mathbf{x}) \, ds, \quad p = 1, 2, \quad i = 1, 2, 3,
 \end{aligned}
 \tag{70}$$

where  $\sigma_{ij}$  is the stress calculated from displacement  $\mathbf{v}$ . Using Eq. (68) we then find

$$\begin{aligned}
 \frac{F_1^{\text{el}(1,2)}}{16\pi\mu a^2(1-\nu)} &= [(t_1^{(2)} + t_1^{(1)})B_0^{\eta_1} \pm (t_1^{(2)} - t_1^{(1)})B_0^{\zeta_1} \\
 &\quad + (\omega_2^{(2)} - \omega_2^{(1)})B_0^{\zeta_5} \pm (\omega_2^{(2)} + \omega_2^{(1)})B_0^{\eta_5}], \\
 \frac{F_2^{\text{el}(1,2)}}{16\pi\mu a^2(1-\nu)} &= [(t_2^{(2)} + t_2^{(1)})B_0^{\eta_1} \pm (t_2^{(2)} - t_2^{(1)})B_0^{\zeta_1} \\
 &\quad - (\omega_1^{(2)} - \omega_1^{(1)})B_0^{\zeta_5} \pm (\omega_1^{(2)} + \omega_1^{(1)})B_0^{\eta_5}], \\
 \frac{F_3^{\text{el}(1,2)}}{16\pi\mu a^2(1-\nu)} &= [(t_3^{(2)} + t_3^{(1)})B_0^{\eta_3} \pm (t_3^{(2)} - t_3^{(1)})B_0^{\zeta_3}].
 \end{aligned}
 \tag{71}$$

The coefficients  $B_0^{\zeta_1} \dots B_0^{\eta_3}$  in Eq. (71) depend on  $\nu$  and the ratio  $a/r$ , where  $r$  is the distance between  $\Omega_1$  and  $\Omega_2$  in the unperturbed state; explicit expressions for these coefficients are given in Appendix A. Similarly, the torque acting on particle  $\Omega_p$  is given by

$$\begin{aligned}
 \mathcal{T}_i^{\text{el}(p)} &= a\varepsilon_{ijk} \int_{\partial\Omega_p} n_j^{(p)}(\mathbf{x})[\sigma_{km}(\mathbf{x}) + \tilde{\sigma}_{km}(\mathbf{x})]n_m^{(p)}(\mathbf{x}) \, ds \\
 &= a\varepsilon_{ijk} \int_{\partial\Omega_p} n_j^{(p)}(\mathbf{x})\sigma_{km}(\mathbf{x})n_m^{(p)}(\mathbf{x}) \, ds,
 \end{aligned}
 \tag{72}$$

or, equivalently,

$$\begin{aligned}
 \frac{\mathcal{T}_1^{\text{el}(1,2)}}{16\pi\mu a^2(1-\nu)} &= [- (t_2^{(2)} - t_2^{(1)})(B_1^{\zeta_1} + C_1^{\zeta_1}) \mp (t_2^{(2)} + t_2^{(1)})(B_1^{\eta_1} + C_1^{\eta_1}) \\
 &\quad \pm (\omega_1^{(2)} - \omega_1^{(1)})(B_1^{\zeta_5} + C_1^{\zeta_5}) + (\omega_1^{(2)} + \omega_1^{(1)})(B_1^{\eta_5} + C_1^{\eta_5})], \\
 \frac{\mathcal{T}_2^{\text{el}(1,2)}}{16\pi\mu a^2(1-\nu)} &= [(t_1^{(2)} - t_1^{(1)})(B_1^{\zeta_1} + C_1^{\zeta_1}) \pm (t_1^{(2)} + t_1^{(1)})(B_1^{\eta_1} + C_1^{\eta_1}) \\
 &\quad \pm (\omega_2^{(2)} - \omega_2^{(1)})(B_1^{\zeta_5} + C_1^{\zeta_5}) + (\omega_2^{(2)} + \omega_2^{(1)})(B_1^{\eta_5} + C_1^{\eta_5})], \\
 \frac{\mathcal{T}_3^{\text{el}(1,2)}}{16\pi\mu a^2(1-\nu)} &= [\pm (\omega_3^{(2)} - \omega_3^{(1)})B_1^{\zeta_6} + (\omega_3^{(2)} + \omega_3^{(1)})B_1^{\eta_6}].
 \end{aligned}
 \tag{73}$$

**4.2. Magnetic interaction**

Consider the reference pair of particles  $\Omega_1$  and  $\Omega_2$ , and assume that particle  $\Omega_p$  has undergone a rigid body translation  $a\mathbf{T}^{(p)}$ , for  $p = 1, 2$ , see Eq. (61). The interparticle distance is given by

$$|\mathbf{x}^{(2)} - \mathbf{x}^{(1)}| = a\sqrt{\left(T_1^{(2)} - T_1^{(1)}\right)^2 + \left(T_2^{(2)} - T_2^{(1)}\right)^2 + \left(\frac{r}{a} + T_3^{(2)} - T_3^{(1)}\right)^2}, \quad (74)$$

where  $r$  is the distance between the particles in the unperturbed state. (Naturally, the particle motions are assumed to be infinitesimal, to accommodate our assumptions of linear elasticity and our use of the isotropic two-point probability density function (49) for the particle configuration after deformation.) As  $\Omega_1$  and  $\Omega_2$  are translated by  $T_j^{(1,2)}$ , the axis of their centers rotates with respect to its initial direction  $\mathbf{e}_3$ ; in what follows we call  $\mathbf{e}'_1, \mathbf{e}'_2$  and  $\mathbf{e}'_3$  the unit vectors of the rotated system of coordinates, where  $\mathbf{e}'_3$  is taken along  $\mathbf{x}^{(2)} - \mathbf{x}^{(1)}$  and points from  $\Omega_1$  towards  $\Omega_2$ . The direction cosines of  $\mathbf{e}'_3$  with respect to the unperturbed coordinate axes  $\mathbf{e}_1, \mathbf{e}_2$  and  $\mathbf{e}_3$  are given by

$$\begin{aligned} \cos \vartheta_1 &= \frac{a(T_1^{(2)} - T_1^{(1)})}{|\mathbf{x}^{(2)} - \mathbf{x}^{(1)}|}, \\ \cos \vartheta_2 &= \frac{a(T_2^{(2)} - T_2^{(1)})}{|\mathbf{x}^{(2)} - \mathbf{x}^{(1)}|}, \\ \cos \vartheta_3 &= \frac{r + a(T_3^{(2)} - T_3^{(1)})}{|\mathbf{x}^{(2)} - \mathbf{x}^{(1)}|}. \end{aligned} \quad (75)$$

The forces of magnetic interaction between particles  $\Omega_1$  and  $\Omega_2$  may be calculated either as (see Jackson (1975))  $\mathbf{F}^{\text{mag}(1)} = \int_{\partial\Omega_1} [\mathbf{M}^{(1)} \cdot \mathbf{n}^{(1)}(\mathbf{x})]\mathbf{H}^{(2)}(\mathbf{x}) \, ds$  or, as

$$\mathbf{F}^{\text{mag}(1)} = -\frac{1}{a}\nabla_{T^{(1)}}\mathcal{W}_{1,2}^{\text{mag}} = \frac{1}{a}\nabla_{T^{(2)}}\mathcal{W}_{1,2}^{\text{mag}} = -\mathbf{F}^{\text{mag}(2)}, \quad (76)$$

where

$$\mathcal{W}_{1,2}^{\text{mag}} = \mathcal{W}^{\text{mag}(1)} + \mathcal{W}^{\text{mag}(2)}. \quad (77)$$

Of course, the results of these two calculations coincide, and the net magnetic force acting on particle  $\Omega^{(1)}$  results:

$$\begin{aligned} \mathbf{F}^{\text{mag}(1)} \cdot \mathbf{e}'_1 &= -\frac{16\pi^2}{3}a^2 \left(\frac{a}{|\mathbf{x}^{(2)} - \mathbf{x}^{(1)}|}\right)^4 [(\mathbf{M}^{(1)} \cdot \mathbf{e}'_1)(\mathbf{M}^{(2)} \cdot \mathbf{e}'_3) \\ &\quad + (\mathbf{M}^{(1)} \cdot \mathbf{e}'_3)(\mathbf{M}^{(2)} \cdot \mathbf{e}'_1)], \\ \mathbf{F}^{\text{mag}(1)} \cdot \mathbf{e}'_2 &= -\frac{16\pi^2}{3}a^2 \left(\frac{a}{|\mathbf{x}^{(2)} - \mathbf{x}^{(1)}|}\right)^4 [(\mathbf{M}^{(1)} \cdot \mathbf{e}'_2)(\mathbf{M}^{(2)} \cdot \mathbf{e}'_3) \\ &\quad + (\mathbf{M}^{(1)} \cdot \mathbf{e}'_3)(\mathbf{M}^{(2)} \cdot \mathbf{e}'_2)], \end{aligned}$$

$$\mathbf{F}^{\text{mag}(1)} \cdot \mathbf{e}'_3 = -\frac{16\pi^2}{3} a^2 \left( \frac{a}{|\mathbf{x}^{(2)} - \mathbf{x}^{(1)}|} \right)^4 \times [\mathbf{M}^{(1)} \cdot \mathbf{M}^{(2)} - 3(\mathbf{M}^{(1)} \cdot \mathbf{e}'_3)(\mathbf{M}^{(2)} \cdot \mathbf{e}'_3)]. \tag{78}$$

We note that, under our present assumption of a vanishing magnetic anisotropy energy, the torques induced by magnetic interactions vanish as well.

### 4.3. Magneto-elastic equilibrium

We wish to calculate  $t_i^{(p)}$  and  $\omega_i^{(p)}$ ,  $i = 1, 2, 3$ ,  $p = 1, 2$ , (see Eq. (66)) from the conditions that, at equilibrium, the elastic and magnetic forces and torques balance each other. Bearing in mind that, as pointed out above, there are no magnetic torques in our context, from Eqs. (71) and (73) we obtain

$$\begin{aligned} t_i^{(2)} &= -t_i^{(1)}, \quad i = 1, 2, 3, \\ \omega_1^{(2)} &= \omega_1^{(1)} = -t_2^{(1)} \frac{B_1^{\chi_1} + C_1^{\chi_1}}{B_1^{\eta_5} + C_1^{\eta_5}}, \\ \omega_2^{(2)} &= \omega_2^{(1)} = t_1^{(1)} \frac{B_1^{\chi_1} + C_1^{\chi_1}}{B_1^{\eta_5} + C_1^{\eta_5}}, \\ \omega_3^{(2)} &= \omega_3^{(1)} = 0 \end{aligned} \tag{79}$$

and the elastic force acting on  $\Omega_1$  takes the simple form

$$\mathbf{F}^{\text{el}(1)} = -32\pi\mu a^2(1 - \nu)\mathcal{D}\mathbf{t} \tag{80}$$

where  $\mathbf{t} = \begin{pmatrix} t_1^{(1)} \\ t_2^{(1)} \\ t_3^{(1)} \end{pmatrix}$  and  $\mathcal{D}$  is the positive diagonal matrix (see Eq. (71))

$$\mathcal{D} = \text{diag} \left\{ B_0^{\chi_1} - B_0^{\eta_5} \frac{B_1^{\chi_1} + C_1^{\chi_1}}{B_1^{\eta_5} + C_1^{\eta_5}}, B_0^{\chi_1} - B_0^{\eta_5} \frac{B_1^{\chi_1} + C_1^{\chi_1}}{B_1^{\eta_5} + C_1^{\eta_5}}, B_0^{\chi_3} \right\}. \tag{81}$$

The magnetizations  $\mathbf{M}^{(1)}$  and  $\mathbf{M}^{(2)}$  of particles  $\Omega_1$  and  $\Omega_2$ , in turn, are minimizers of the energy  $\mathcal{W}_{1,2}^{\text{mag}}$ . The condition of minimization, in fact, can be used to eliminate one of the two magnetizations in favor of the other. Indeed, suppose first that the magnetizations of the particles  $\Omega_1$  and  $\Omega_2$  are below saturation. Then the magnetizations satisfy the optimality conditions

$$\begin{aligned} \frac{16\pi^2 a^3}{9} \left\{ \mathbf{M}^{(1)} + \left( \frac{a}{|\mathbf{x}^{(2)} - \mathbf{x}^{(1)}|} \right)^3 [\mathbf{M}^{(2)} - 3\mathbf{e}'_3(\mathbf{M}^{(2)} \cdot \mathbf{e}'_3)] \right\} &= \frac{4\pi a^3}{3} \mathbf{H}_0 \\ \frac{16\pi^2 a^3}{9} \left\{ \mathbf{M}^{(2)} + \left( \frac{a}{|\mathbf{x}^{(2)} - \mathbf{x}^{(1)}|} \right)^3 [\mathbf{M}^{(1)} - 3\mathbf{e}'_3(\mathbf{M}^{(1)} \cdot \mathbf{e}'_3)] \right\} &= \frac{4\pi a^3}{3} \mathbf{H}_0. \end{aligned} \tag{82}$$

It is a matter of simple algebra to show that Eq. (82) implies  $\mathbf{M}^{(1)} = \mathbf{M}^{(2)}$ . At very high applied magnetic fields  $\mathbf{H}_0$ , the magnetization saturates and the constraints

$$|\mathbf{M}^{(p)}| \leq M^{\text{sat}}, \quad p = 1, 2, \tag{83}$$

become active. The analysis of this case is given in Appendix B; there we show that, at saturation, the minimizing magnetizations satisfy  $\mathbf{M}^{(1)} = \mathbf{M}^{(2)}$ , as well.

The problem of mechanical equilibrium thus becomes: Find  $\mathbf{t}$  and  $\mathbf{M}^{(1)}$  such that  $\mathbf{F}^{\text{el}(1)} + \mathbf{F}^{\text{mag}(1)} = 0$ . This is a coupled magneto-elastic problem which we solve by minimizing the total energy

$$\begin{aligned} \mathcal{W}_{1,2} = & 32\pi\mu a^3(1-\nu)\mathbf{t} \cdot \mathcal{D}\mathbf{t} - \frac{8\pi}{3}a^3\mathbf{M}^{(1)} \cdot \mathbf{H}_0 + \frac{16\pi^2}{9}a^3|\mathbf{M}^{(1)}|^2 \\ & + \frac{16\pi^2}{9}a^3 \left( \frac{a}{|\mathbf{x}^{(2)} - \mathbf{x}^{(1)}|} \right)^3 [|\mathbf{M}^{(1)}|^2 - 3(\mathbf{M}^{(1)} \cdot \mathbf{e}_3')^2], \end{aligned} \quad (84)$$

over the magnetization  $\mathbf{M}^{(1)}$  and translations  $\mathbf{t}$ , subject to constraints (83) and  $|\mathbf{x}^{(2)} - \mathbf{x}^{(1)}| \geq 2a$ . Finally, for a given strain  $\varepsilon^0$ , displacements  $\tilde{t}_j^{(p)}, \tilde{\omega}_j^{(p)}, j = 1, 2, 3$  and  $p = 1, 2$  in Eqs. (64) and (66) are calculated with the formulas given by Chen and Acrivos (1978a).

## 5. Overall magneto-elastic behavior

In the following three sections, we write the potential energy  $\mathcal{U}$  in a convenient form for numerical calculations. We utilize the solutions of magnetic and elastic problems developed in the previous sections to describe the overall magneto-elastic behavior of the composites under consideration. In the absence of an external magnetic field the problem reduces to homogenization in linear elasticity (Bensoussan et al., 1978; Jikov et al., 1994) [21] and the macroscopic properties of the composite are completely determined by the average stress. However, when a magnetic field  $\mathbf{H}_0$  is applied, the average stress gives only a part of the strain energy (see Eq. (27)) and it is not a physical observable. Thus, instead of evaluating average stresses, we focus on obtaining the average strain in the composite, for given surface tractions at  $\partial V$  and given applied magnetic fields. The main tool in our calculation is the variational principle described in Remark 2: Given boundary tractions

$$S_i(\mathbf{x}) = \eta_{ij}^0 n_j(\mathbf{x}), \quad \mathbf{x} \in \partial V, \quad (85)$$

with constant  $\eta_{ij}^0$ , and due to the uniform distribution of particles in  $V$ , the displacement at the boundary is homogeneous

$$u_i(\mathbf{x}) = \varepsilon_{ij}^0 x_j, \quad \mathbf{x} \in \partial V, \quad i = 1, 2, 3. \quad (86)$$

We thus can obtain such an homogeneous deformation of  $V$  as the minimizer  $\varepsilon_{ij}^0$  of the potential energy  $\mathcal{U}$  over all homogeneous strains.

### 5.1. The elastic energy

The displacement  $\mathbf{u}(\mathbf{x})$  is given by superposition (65)

$$\mathbf{u}(\mathbf{x}) = \tilde{\mathbf{v}}(\mathbf{x}) + \mathbf{v}(\mathbf{x}), \quad (87)$$

where  $\tilde{\mathbf{v}}$  solves the pure elasticity problem (no body forces) in  $V$  and

$$\begin{aligned} \tilde{\mathbf{v}}(\mathbf{x}) &= \mathbf{u}(\mathbf{x}) = \varepsilon_{ij}^0 x_j && \text{for } \mathbf{x} \in \partial V, \\ \tilde{\mathbf{v}}(\mathbf{x}) &= a\tilde{t}_i^{(p)} \mathbf{e}_i + a\omega_j^{(p)} \varepsilon_{ijk} n_k^{(p)}(\mathbf{x}) \mathbf{e}_i && \text{for } \mathbf{x} \in \partial\Omega_p, \quad p = 1, \dots, N. \end{aligned} \tag{88}$$

The displacement  $\mathbf{v}(\mathbf{x})$  in Eq. (87) is due to magnetic interactions in  $V$  and it satisfies boundary conditions

$$\begin{aligned} \mathbf{v}(\mathbf{x}) &= \mathbf{0} && \text{for } \mathbf{x} \in \partial V, \\ \mathbf{v}(\mathbf{x}) &= a\tilde{t}_i^{(p)} \mathbf{e}_i + a\omega_j^{(p)} \varepsilon_{ijk} n_k^{(p)}(\mathbf{x}) \mathbf{e}_i && \text{for } \mathbf{x} \in \partial\Omega_p, \quad p = 1, \dots, N. \end{aligned} \tag{89}$$

Consistent with Eq. (87), the stress and strain are

$$\begin{aligned} \Sigma_{ij}(\mathbf{x}) &= \tilde{\sigma}_{ij}(\mathbf{x}) + \sigma_{ij}(\mathbf{x}), \\ E_{ij}(\mathbf{x}) &= \tilde{\varepsilon}_{ij}(\mathbf{x}) + \varepsilon_{ij}(\mathbf{x}), \end{aligned} \tag{90}$$

where, for  $\mathbf{x}$  in the elastic matrix,

$$\begin{aligned} \tilde{\varepsilon}_{ij}(\mathbf{x}) &= \frac{1}{2} \left( \frac{\partial \tilde{v}_i}{\partial x_j} + \frac{\partial \tilde{v}_j}{\partial x_i} \right), \\ \varepsilon_{ij}(\mathbf{x}) &= \frac{1}{2} \left( \frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right), \\ \frac{\partial \tilde{\sigma}_{ij}(\mathbf{x})}{\partial x_j} &= \frac{\partial \sigma_{ij}(\mathbf{x})}{\partial x_j} = 0 \quad \text{for } i = 1, 2, 3, \end{aligned} \tag{91}$$

and, for  $p = 1, \dots, N$ ,

$$\begin{aligned} \mathbf{e}_i \int_{\partial\Omega_p} \tilde{\sigma}_{ij}(\mathbf{x}) n_j^{(p)}(\mathbf{x}) \, ds &= 0, \\ \varepsilon_{ijk} \int_{\partial\Omega_p} n_j^{(p)}(\mathbf{x}) \tilde{\sigma}_{km}(\mathbf{x}) n_m^{(p)}(\mathbf{x}) \, ds &= 0, \\ \mathbf{e}_i \int_{\partial\Omega_p} \sigma_{ij}(\mathbf{x}) n_j^{(p)}(\mathbf{x}) \, ds &= \mathbf{F}^{\text{el}(p)} = -\mathbf{F}^{\text{mag}(p)}, \\ a\mathbf{e}_i \varepsilon_{ijk} \int_{\partial\Omega_p} n_j^{(p)}(\mathbf{x}) \sigma_{km}(\mathbf{x}) n_m^{(p)}(\mathbf{x}) \, ds &= \mathcal{F}^{\text{el}(p)} = 0. \end{aligned} \tag{92}$$

Recalling notation  $V_M = V \setminus \bigcup_{p=1}^N \Omega_p$  for the volume occupied functional is

$$\begin{aligned} \langle \mathcal{W}^{\text{el}} \rangle &= \frac{1}{2V} \int_{V_M} \Sigma_{ij}(\mathbf{x}) E_{ij}(\mathbf{x}) \, dx \\ &= \frac{1}{2V} \int_{V_M} [\tilde{\sigma}_{ij}(\mathbf{x}) \tilde{\varepsilon}_{ij}(\mathbf{x}) + \sigma_{ij}(\mathbf{x}) \varepsilon_{ij}(\mathbf{x})] \, dx \\ &\quad + \frac{1}{2V} \int_{V_M} [\tilde{\sigma}_{ij}(\mathbf{x}) \varepsilon_{ij}(\mathbf{x}) + \sigma_{ij}(\mathbf{x}) \tilde{\varepsilon}_{ij}(\mathbf{x})] \, dx. \end{aligned} \tag{93}$$

By Hooke’s law, we have

$$\tilde{\sigma}_{ij}(\mathbf{x})\varepsilon_{ij}(\mathbf{x}) = [\lambda\tilde{\varepsilon}_{kk}(\mathbf{x})\delta_{ij} + 2\mu\tilde{\varepsilon}_{ij}(\mathbf{x})]\varepsilon_{ij}(\mathbf{x}) = \sigma_{ij}(\mathbf{x})\tilde{\varepsilon}_{ij}(\mathbf{x})$$

so that Eqs. (89) and (92) give

$$\begin{aligned} & \frac{1}{2V} \int_{V_M} [\tilde{\sigma}_{ij}(\mathbf{x})\varepsilon_{ij}(\mathbf{x}) + \sigma_{ij}(\mathbf{x})\tilde{\varepsilon}_{ij}(\mathbf{x})] \, d\mathbf{x} \\ &= \frac{1}{V} \int_{V_M} \tilde{\sigma}_{ij}(\mathbf{x})\varepsilon_{ij}(\mathbf{x}) \, d\mathbf{x} = \frac{1}{V} \int_{V_M} \tilde{\sigma}_{ij}(\mathbf{x}) \frac{\partial v_i(\mathbf{x})}{\partial x_j} \, d\mathbf{x} \\ &= \frac{1}{V} \int_{\partial V} v_i(\mathbf{x})\tilde{\sigma}_{ij}(\mathbf{x})n_j(\mathbf{x}) \, ds - \frac{1}{V} \sum_{p=1}^N \int_{\partial\Omega_p} v_i(\mathbf{x})\tilde{\sigma}_{ij}(\mathbf{x})n_j^{(p)}(\mathbf{x}) \, ds = 0. \end{aligned} \tag{94}$$

Energy term

$$\frac{1}{2V} \int_{V_M} \tilde{\sigma}_{ij}(\mathbf{x})\tilde{\varepsilon}_{ij}(\mathbf{x}) \, d\mathbf{x} = \frac{1}{2} \varepsilon_{ij}^0 \langle \tilde{\sigma}_{ij} \rangle = \lambda^\star (\varepsilon_{kk}^0)^2 + \mu^\star \varepsilon_{ij}^0 \varepsilon_{ij}^0, \tag{95}$$

corresponds to the pure elasticity problem (absence of a magnetic field) and the effective Lamé constant  $\lambda^\star$  and shear modulus  $\mu^\star$  were evaluated to  $O(\Phi^2)$  in Chen and Acrivos (1978b). Finally, Eqs. (91), (92) and (89) give

$$\begin{aligned} & \frac{1}{2V} \int_{V_M} \sigma_{ij}(\mathbf{x})\varepsilon_{ij}(\mathbf{x}) \, d\mathbf{x} \\ &= \frac{1}{2V} \int_{V_M} \sigma_{ij}(\mathbf{x}) \frac{\partial v_i(\mathbf{x})}{\partial x_j} \, d\mathbf{x} = \frac{1}{2V} \int_{V_M} \frac{\partial [v_i(\mathbf{x})\sigma_{ij}(\mathbf{x})]}{\partial x_j} \, d\mathbf{x} \\ &= -\frac{1}{2V} \sum_{p=1}^N \int_{\partial\Omega_p} \sigma_{ij}(\mathbf{x})v_i(\mathbf{x})n_j^{(p)}(\mathbf{x}) \, ds \\ &= -\frac{1}{2V} \sum_{p=1}^N [at_i^{(p)}F_i^{\text{el}(p)} + \omega_i^{(p)}\mathcal{F}_i^{\text{el}(p)}] \end{aligned} \tag{96}$$

and, since the net torque  $\mathcal{F}^{\text{el}(p)}$  is zero (see Section 4.2), we have

$$\frac{1}{2V} \int_{V_M} \sigma_{ij}(\mathbf{x})\varepsilon_{ij}(\mathbf{x}) \, d\mathbf{x} = -\frac{\mathcal{N}}{2} \int_{r \geq 2a} at_i^{(1)}F_i^{\text{el}(1)}P(\mathbf{r}|0) \, d\mathbf{r}. \tag{97}$$

Note that quantity  $\mathbf{t}$  in Eq. (97) decays like  $1/r^4$  as  $r \rightarrow \infty$ , and, therefore, the integral is absolutely convergent.

### 5.2. The magnetic energy

The magnetic energy is given by

$$\langle \mathcal{W}^{\text{mag}} \rangle = \mathcal{N} \overline{\mathcal{W}^{\text{mag}(1)}}, \tag{98}$$

where we average  $\mathcal{W}^{\text{mag}(1)}$  given by Eq. (57), over the particles. When the distance between  $\Omega_1$  and all other particles in  $V$  is much greater than  $a$ , the reference particle is essentially isolated in the matrix and its magnetization is  $\mathbf{M}^\star$ , given by Eq. (60). Thus, we rewrite Eq. (98) as

$$\begin{aligned} \langle \mathcal{W}^{\text{mag}} \rangle = \mathcal{N} & \left[ -\frac{4\pi a^3}{3} \mathbf{M}^\star \mathbf{H}_0 + \frac{8\pi^2 a^3}{9} (\mathbf{M}^\star)^2 \right] \\ & + \mathcal{N} \int_{r \geq 2a} P(\mathbf{r}|0) \left\{ -\frac{4\pi a^3}{3} (\mathbf{M}^{(1)} - \mathbf{M}^\star) \cdot \mathbf{H}_0 \right. \\ & \left. + \frac{8\pi^2 a^3}{9} (|\mathbf{M}^{(1)}|^2 - |\mathbf{M}^\star|^2) \right\} d\mathbf{r} + \mathcal{N} \bar{\mathcal{G}}, \end{aligned} \tag{99}$$

where  $\mathbf{M}^{(1)}$ , the minimizer of Eq. (84), depends on  $\mathbf{r}$  and

$$\mathcal{G} = \frac{8\pi^2 a^3}{9} \left[ \frac{a}{|\mathbf{r}^{\text{eq}}(\mathbf{r}^{(1)})|} \right]^3 [|\mathbf{M}^{(1)}|^2 - 3(\mathbf{M}^{(1)} \cdot \mathbf{e}'_3)^2] \tag{100}$$

(see Eq. (57)). Suppose that  $\Omega_{q(1)}$  is in the vicinity of  $\Omega_1$ , and, in the unperturbed state,  $\Omega_1$  and  $\Omega_{q(1)}$  have the relative vector position  $\mathbf{r}^{(1)}$ . In Eq. (100),  $\mathbf{r}^{\text{eq}}(\mathbf{r}^{(1)})$  stands for  $\mathbf{x}^{(q(1))} - \mathbf{x}^{(1)}$  at mechanical equilibrium, and  $\mathbf{e}'_3 = \mathbf{r}^{\text{eq}}(\mathbf{r}^{(1)})/|\mathbf{r}^{\text{eq}}(\mathbf{r}^{(1)})|$ .

The integral in Eq. (99) considers the reference pair of particles  $\Omega_1$  and  $\Omega_2$ , and  $\mathbf{r}$  is the relative vector position between  $\Omega_1$  and  $\Omega_2$ , at zero boundary tractions and no applied magnetic field. We show next that this integral is absolutely convergent. We start with the case of magnetic fields  $\mathbf{H}_0$  that are not sufficiently strong to achieve saturation, such that  $\mathbf{M}^{(1)}$  satisfies the first order optimality condition (Gill et al., 1989)

$$\begin{aligned} \frac{1}{2} \nabla_{\mathbf{M}^{(1)}} \mathcal{W}_{1,2}^{\text{mag}} & = -\frac{4\pi a^3}{3} \mathbf{H}_0 + \frac{16\pi^2 a^3}{9} \mathbf{M}^{(1)} \\ & + \frac{16\pi^2 a^3}{9} \left( \frac{a}{|\mathbf{r}^{\text{eq}}|} \right)^3 [\mathbf{M} - 3\mathbf{e}'_3(\mathbf{M} \cdot \mathbf{e}'_3)] = \mathbf{0}. \end{aligned} \tag{101}$$

For  $r \gg a$ ,  $\mathbf{M}^{(1)} = \mathbf{M}^\star + \delta\mathbf{M}$ , where  $\mathbf{M}^\star = 3/4\pi\mathbf{H}_0$ . Then, from Eq. (101), we have

$$\delta\mathbf{M} + \left( \frac{a}{|\mathbf{r}^{\text{eq}}|} \right)^3 [\mathbf{M}^\star + \delta\mathbf{M} - 3\mathbf{e}'_3(\mathbf{M}^\star + \delta\mathbf{M}) \cdot \mathbf{e}'_3] = \mathbf{0} \tag{102}$$

or, equivalently,

$$\delta\mathbf{M} = - \left( \frac{a}{|\mathbf{r}^{\text{eq}}|} \right)^3 M^\star [\zeta_3 - 3\mathbf{e}'_3(\zeta_3 \cdot \mathbf{e}'_3)] + O\left(\frac{a}{r}\right)^6. \tag{103}$$

Hence,

$$-\frac{3}{2\pi} (\mathbf{M}^{(1)} - \mathbf{M}^\star) \cdot \mathbf{H}_0 + (|\mathbf{M}^{(1)}|^2 - |\mathbf{M}^\star|^2) = O\left(\frac{a}{r}\right)^6 \tag{104}$$

and its integral is absolutely convergent. Next, suppose that  $\mathbf{M}^{(1)}$  is saturated. Since  $\mathbf{M}^{(1)}$  is the minimizer of  $\mathcal{W}_{1,2}^{\text{mag}}$ , it must satisfy the first order optimality condition (Gill et al., 1989)

$$\nabla_{\mathbf{M}^{(1)}} \mathcal{W}_{1,2}^{\text{mag}} \cdot \mathbf{p} = \mathbf{0} \quad \text{where } \mathbf{M}^{(1)} \cdot \mathbf{p} = 0. \tag{105}$$

Clearly, for  $r/a \rightarrow \infty$ ,  $\mathbf{M}^{(1)} = \mathbf{M}^\star = M^\star \zeta_3 = M^{\text{sat}} \zeta_3$ . Consider  $\mathbf{p}^\star$ , a unit vector chosen in the plane defined by  $\mathbf{M}^{(1)}$  and  $\zeta_3$ , where  $\zeta_3 \cdot \mathbf{p}^\star = 0$ . Thus,

$$\mathbf{p}^\star = \cos \beta \zeta_1 + \sin \beta \zeta_2, \quad (106)$$

for some angle  $\beta$ . Let  $\delta \ll 1$  be the angle between  $\mathbf{M}^{(1)}$  and  $\zeta_3$ , for  $r \gg a$ . Then,

$$\mathbf{M}^{(1)} = M^\star (\cos \delta \zeta_3 + \sin \delta \mathbf{p}^\star). \quad (107)$$

Now take  $\mathbf{p} = \cos \delta \mathbf{p}^\star - \sin \delta \zeta_3$ , which is clearly orthogonal to  $\mathbf{M}^{(1)}$ . From the optimality condition  $\nabla_{\mathbf{M}^{(1)}} \mathcal{W}_{1,2}^{\text{mag}} \cdot \mathbf{p} = 0$ , we obtain

$$\frac{H_0}{4\pi} \sin \delta = \left( \frac{a}{|\mathbf{r}^{\text{eq}}|} \right)^3 (\mathbf{p} \cdot \mathbf{e}'_3)(\mathbf{M}^{(1)} \cdot \mathbf{e}'_3) \quad (108)$$

and, from the definition of  $\mathbf{p}$ , Eqs. (106) (107) and  $\delta \ll 1$ , we find

$$\delta \approx \frac{4\pi M^\star}{H_0} \left( \frac{a}{|\mathbf{r}^{\text{eq}}|} \right)^3 [\cos \beta (\zeta_1 \cdot \mathbf{e}'_3)(\zeta_3 \cdot \mathbf{e}'_3) + \sin \beta (\zeta_2 \cdot \mathbf{e}'_3)(\zeta_3 \cdot \mathbf{e}'_3)]. \quad (109)$$

Hence,

$$-(\mathbf{M}^{(1)} - \mathbf{M}^\star) \cdot \mathbf{H}_0 = M^\star H_0 (1 - \cos \delta) \approx \frac{1}{2} M^\star H_0 \delta^2 = O\left(\frac{a}{r}\right)^6$$

and its integral is absolutely convergent.

### 5.3. An explicit expression of the overall energy functional

We gather the results of Sections 5.1 and 5.2; since  $\mathbf{M}^\star$  is constant, we replace the functional  $\mathcal{U}$  in Eq. (19) by  $\mathcal{U}' = \mathcal{U} - \mathcal{N}[-4\pi a^3/3M^\star H_0 + 8\pi^2 a^3/9(M^\star)^2]$ . Using spherical coordinates  $r$ ,  $\theta$  and  $\phi$  and a change of variables  $\rho = a/r$  we obtain

$$\begin{aligned} \mathcal{U}' &= \frac{\lambda^\star}{2} (\varepsilon_{kk}^0)^2 + \mu^\star \varepsilon_{ij}^0 \varepsilon_{ij}^0 - \varepsilon_{ij}^0 \eta_{ij}^0 + \frac{\Phi^2}{2} \int_0^{\frac{1}{2}} d\rho \int_0^\pi d\theta \int_0^{2\pi} d\phi \frac{\sin \theta}{\rho^4} \\ &\times \left\{ \frac{18}{\pi} \mu(1-\nu) \mathbf{t} \cdot \mathcal{D} \mathbf{t} - \frac{3}{2\pi} (\mathbf{M}^{(1)} - \mathbf{M}^\star) \cdot \mathbf{H}_0 + (|\mathbf{M}^{(1)}|^2 - |\mathbf{M}^\star|^2) \right\} + \mathcal{N} \overline{\mathcal{G}}. \end{aligned} \quad (110)$$

Here, we have used Eq. (80) for the elastic force and expressions (85), (86) and (49) for the boundary tractions, strain and the probability density  $P(\mathbf{r}|\mathbf{0})$ , respectively. Next, we concentrate on the calculation of (see Eq. (100))

$$\overline{\mathcal{G}} = \frac{1}{N} \sum_{p=1}^N \frac{8\pi^2 a^3}{9} \left( \frac{a}{|\mathbf{r}^{\text{eq}}(\mathbf{r}^{(p)})|} \right)^3 \left[ |\mathbf{M}^{(p)}|^2 - 3 \left( \mathbf{M}^{(p)} \cdot \frac{\mathbf{r}^{\text{eq}}(\mathbf{r}^{(p)})}{|\mathbf{r}^{\text{eq}}(\mathbf{r}^{(p)})|} \right)^2 \right]. \quad (111)$$

Here,  $\mathbf{r}^{(p)}$  is the distance between neighbors  $\Omega_p$  and  $\Omega_{q(p)}$ , at zero strain and no applied magnetic field. Due to the coupled magnetic and elastic interactions, the particles move and, at equilibrium, they are separated by  $\mathbf{r}^{\text{eq}}(\mathbf{r}^{(p)})$ . We cannot write  $\overline{\mathcal{G}}$  as an integral because of the slow, like  $1/r^3$ , decay of the terms in sum (111). Instead, we add and subtract from  $\overline{\mathcal{G}}$  terms that have the same asymptotic behavior at

$|\mathbf{r}^{(p)}/a| \rightarrow \infty$  as those in Eq. (111). For  $|\mathbf{r}^{(p)}| \gg a$ , the particles are isolated and, as we already established, their magnetization is  $\mathbf{M}^{(p)} \approx \mathbf{M}^\star$ . Moreover,  $\mathbf{r}^{\text{eq}}(\mathbf{r}^{(p)}) \approx \mathbf{r}^\star(\mathbf{r}^{(p)})$ , the displacement of an isolated particle, located at  $\mathbf{r}^{(p)}$  with respect to the origin, in the unperturbed state of zero strain. Then,

$$\begin{aligned} \mathcal{N}\overline{\mathcal{G}} &= \frac{\Phi^2}{2} \int_0^{\frac{1}{2}} d\rho \int_0^\pi d\theta \int_0^{2\pi} d\phi \frac{\sin \theta}{\rho^4} \\ &\times \left\{ \left( \frac{a}{|\mathbf{r}^{\text{eq}}(\mathbf{r})|} \right)^3 \left[ |\mathbf{M}^{(1)}|^2 - 3 \left( \mathbf{M}^{(1)} \cdot \frac{\mathbf{r}^{\text{eq}}(\mathbf{r})}{|\mathbf{r}^{\text{eq}}(\mathbf{r})|} \right)^2 \right] \right. \\ &\left. - \left( \frac{a}{|\mathbf{r}^\star(\mathbf{r})|} \right)^3 \left[ |\mathbf{M}^\star|^2 - 3 \left( \mathbf{M}^\star \cdot \frac{\mathbf{r}^\star(\mathbf{r})}{|\mathbf{r}^\star(\mathbf{r})|} \right)^2 \right] \right\} + \mathcal{N}\overline{\Delta\mathcal{G}}, \end{aligned} \tag{112}$$

where  $\rho = a/|\mathbf{r}|$  and

$$\mathcal{N}\overline{\Delta\mathcal{G}} = \Phi \frac{2\pi}{3} \frac{1}{N} \sum_{p=1}^N \left( \frac{a}{|\mathbf{r}^\star(\mathbf{r}^{(p)})|} \right)^3 \left[ |\mathbf{M}^\star|^2 - 3 \left( \mathbf{M}^\star \cdot \frac{\mathbf{r}^\star(\mathbf{r}^{(p)})}{|\mathbf{r}^\star(\mathbf{r}^{(p)})|} \right)^2 \right]. \tag{113}$$

Clearly, integral (112) is absolutely convergent. Moreover, by the law of large numbers, the sum in Eq. (113) converges. We can calculate Eq. (113) numerically by taking  $N \gg 1$  uniformly distributed random locations  $\mathbf{r}^{(p)}$  such that  $|\mathbf{r}^{(p)}| \geq 2a$ . Note however that Eq. (113) can also be obtained as follows. Our goal is the calculation of the energy for the given strain  $\underline{\underline{\varepsilon}}^0$  so let us consider a one-dimensional path  $\gamma(s)$  of variation of  $\underline{\underline{\varepsilon}}^0$  such that  $\gamma(0) = \underline{\underline{0}}$  and  $\gamma(1) = \underline{\underline{\varepsilon}}^0$ . Since  $\mathbf{M}^\star$  is independent of  $\underline{\underline{\varepsilon}}^0$ , the only quantity that changes in Eq. (113) is  $\mathbf{r}^\star$ , by an amount  $\delta\mathbf{r}^\star$ . We have

$$\begin{aligned} \mathcal{N}\delta\overline{\Delta\mathcal{G}} &= -2\pi\Phi \frac{1}{N} \sum_{p=1}^N \left( \frac{a}{|\mathbf{r}^\star(\mathbf{r}^{(p)})|} \right)^4 \left\{ \frac{\mathbf{r}^\star}{|\mathbf{r}^\star|} \left[ |\mathbf{M}^\star|^2 - 3 \left( \mathbf{M}^\star \cdot \frac{\mathbf{r}^\star(\mathbf{r}^{(p)})}{|\mathbf{r}^\star(\mathbf{r}^{(p)})|} \right)^2 \right] \right. \\ &\left. + 2 \frac{\mathbf{M}^\star \cdot \mathbf{r}^\star(\mathbf{r}^{(p)})}{|\mathbf{r}^\star(\mathbf{r}^{(p)})|} \mathbf{M}^\star - 2 \left[ \frac{\mathbf{M}^\star \cdot \mathbf{r}^\star(\mathbf{r}^{(p)})}{|\mathbf{r}^\star(\mathbf{r}^{(p)})|} \right]^2 \frac{\mathbf{r}^\star}{|\mathbf{r}^\star|} \right\} \cdot \frac{\delta\mathbf{r}^\star(\mathbf{r}^{(p)})}{a}. \end{aligned} \tag{114}$$

As  $|\mathbf{r}^{(p)}/a| \rightarrow \infty$ , the terms in Eq. (114) decay as  $(a/|\mathbf{r}^{(p)}|)^4$  and  $\mathcal{N}\delta\overline{\Delta\mathcal{G}}$  can be written as an absolutely convergent volume integral. To calculate Eq. (113), we integrate with respect to  $s$  along the path  $\gamma(s)$  of variation of  $\underline{\underline{\varepsilon}}^0$  and obtain

$$\begin{aligned} \mathcal{N}\overline{\Delta\mathcal{G}} &= \frac{\Phi^2}{2} \int_0^{\frac{1}{2}} d\rho \int_0^\pi d\theta \int_0^{2\pi} d\phi \frac{\sin \theta}{\rho^4} \left( \frac{a}{|\mathbf{r}^\star(\mathbf{r})|} \right)^3 \\ &\times \left[ |\mathbf{M}^\star|^2 - 3 \left( \mathbf{M}^\star \cdot \frac{\mathbf{r}^\star(\mathbf{r})}{|\mathbf{r}^\star(\mathbf{r})|} \right)^2 \right] + C, \end{aligned} \tag{115}$$

where  $C$  is a constant (independent of  $\underline{\underline{\varepsilon}}^0$ ). Our numerical experiments confirm that the two proposed methods to evaluate Eq. (113) do indeed yield the same result.

Eqs. (110) and (112) give the explicit expressions of the energy functional that we minimize respect to  $\underline{\underline{\varepsilon}}^0$  in our numerical calculations.

## 6. Numerical results

In this section, we present results of our evaluations of average strains for given tractions and applied magnetic fields. For simplicity we restrict our discussion to surface tractions which are symmetric with respect to the magnetic axis  $\zeta_3$ :

$$S_i(\mathbf{x}) = \gamma n_i(\mathbf{x}) + \eta[n_i(\mathbf{x}) - 3n_i(\mathbf{x})\delta_{i3}] \quad \text{for } \mathbf{x} \in \partial V, \quad i = 1, 2, 3, \quad (116)$$

for certain constants  $\gamma$  and  $\eta$ . Our problem then becomes symmetric with respect to the axis  $\zeta_3$  and the associated strains can be expressed in the form

$$\varepsilon_{ij}^0 = \varepsilon \delta_{ij} + \tau(\delta_{ij} - 3\delta_{i3}\delta_{j3}), \quad (117)$$

where the parameters  $\varepsilon$  and  $\tau$  are to be calculated as the minimizers of potential energy  $\mathcal{W}'$ .

In all of our numerical experiments integrals (110) and (112) were evaluated numerically by means of adaptive Simpson quadrature rules (Kincaid and Cheney, 1996). To evaluate the integrands we calculate the magnetization, rotation and translation of the reference sphere by minimizing the energy (84) for pairs of inclusions, for each vector  $\mathbf{r} = |\mathbf{r}|(\sin \theta \cos \phi \zeta_1 + \sin \theta \sin \phi \zeta_2 + \cos \theta \zeta_3)$ ; see Fig. 3. The non-linear constrained minimizer of the energy expression (84) is obtained by means of the software package FFSQP (Zhou et al., 1997). To evaluate integral (115), we use that, for strains of form (117) we have

$$|\mathbf{r}^\star(\mathbf{r})| = |\mathbf{r}|[(1 + \varepsilon + \tau)^2 + 3 \cos^2 \theta (\tau^2 - 2\tau\varepsilon - 2\tau)]^{1/2},$$

$$\left( \mathbf{M}^\star \cdot \frac{\mathbf{r}^\star(\mathbf{r})}{|\mathbf{r}^\star(\mathbf{r})|} \right)^2 = \frac{|\mathbf{M}^\star|^2 (1 + \varepsilon - 2\tau)^2 \cos^2 \theta}{(1 + \varepsilon + \tau)^2 + 3 \cos^2 \theta (\tau^2 - 2\tau\varepsilon - 2\tau)}, \quad (118)$$

where  $\mathbf{r} = |\mathbf{r}|(\sin \theta \cos \phi \zeta_1 + \sin \theta \sin \phi \zeta_2 + \cos \theta \zeta_3)$ . As it can readily be checked, the integral in Eq. (115) equals zero. Finally, we minimize the potential energy  $\mathcal{W}'$  given by Eq. (110), over the components  $\varepsilon$  and  $\tau$  of the strain  $\underline{\underline{\varepsilon}}^0$  (see Eq. (117)), with the MATLAB program *fmins*.

### 6.1. Scaling

Clearly the functional  $\mathcal{W}'/\mu$  is completely determined by the following dimensionless quantities: the strains  $\varepsilon$  and  $\tau$ , the normalized surface tractions  $\gamma/\mu$  and  $\eta/\mu$ , the volume fraction  $\Phi$  and the parameters  $\mathcal{R} = (M^{\text{sat}})^2/2\mu$  and  $\mathcal{Q} = M^{\text{sat}}H_0/2\mu$ . In all of the following numerical experiments we take  $M^{\text{sat}} = 1.91 \times 10^3 \text{ G}$  = the saturation magnetization of an iron–cobalt alloy (Jolly et al., 1996b). The Poisson ratio  $\nu$  is taken in the interval  $(0, 0.5)$  and  $\mu$  varies in the range  $(8 \times 10^5, 3.2 \times 10^6) \text{ dyn/cm}^2$ . The value  $\mu = 3.2 \times 10^6 \text{ dyn/cm}^2$  applies to vulcanized rubber. The applied magnetic field  $H_0$  varies in the range  $(10^3, 10^4) \text{ Oe}$  where a field of  $8.5 \times 10^3 \text{ Oe}$  is high enough to saturate the ferromagnetic particles suspended in the matrix (Jolly et al., 1996a; Morish, 1965). For definiteness, in all numerical experiments below we assume a volume fraction  $\Phi = 0.2$ . Finally, even though the calculations are dimensionless, to illustrate our numerical results we give representative values of the actual parameters used in each numerical experiment.

Table 1  
Self deformation for various applied magnetic fields

$H_0 \times 10^3$ Oe	$\varepsilon$	$\tau$	$\varepsilon_{11}^0 = \varepsilon_{22}^0$	$\varepsilon_{33}^0$
8.119	$-4.466 \times 10^{-2}$	$-2.754 \times 10^{-2}$	$-7.421 \times 10^{-2}$	$8.425 \times 10^{-3}$
7.731	$-3.590 \times 10^{-2}$	$-6.551 \times 10^{-3}$	$-4.245 \times 10^{-2}$	$-2.280 \times 10^{-2}$
7.276	$-3.958 \times 10^{-2}$	$4.700 \times 10^{-3}$	$-3.488 \times 10^{-2}$	$-4.898 \times 10^{-2}$
6.821	$-3.352 \times 10^{-2}$	$5.825 \times 10^{-3}$	$-2.769 \times 10^{-2}$	$-4.517 \times 10^{-2}$
6.367	$-2.706 \times 10^{-2}$	$6.591 \times 10^{-3}$	$-2.047 \times 10^{-2}$	$-4.025 \times 10^{-2}$
5.912	$-1.858 \times 10^{-2}$	$6.638 \times 10^{-3}$	$-1.194 \times 10^{-2}$	$-3.185 \times 10^{-2}$
5.002	$-9.299 \times 10^{-3}$	$-3.342 \times 10^{-5}$	$-9.265 \times 10^{-3}$	$-9.366 \times 10^{-3}$
4.093	$-4.704 \times 10^{-3}$	$-1.439 \times 10^{-3}$	$-6.143 \times 10^{-3}$	$-1.826 \times 10^{-3}$

## 6.2. Self deformation of an elastomer–ferromagnet composite

In our first set of experiments, presented in Table 1, we assume zero tractions on  $\partial V$  and we calculate the deformation of the composite arising from a given applied magnetic field  $H_0 \neq 0$ . We assume that the matrix shear modulus and the Poisson ratio are  $\mu = 8.272 \times 10^5$  dyn/cm<sup>2</sup> ( $= 0.26 \times \mu$  of vulcanized rubber) and  $\nu = 0.4$ , respectively. From Table 1 we note that in all cases considered the material is compressed in the directions  $\zeta_1$  and  $\zeta_2$  orthogonal to the applied field  $\mathbf{H}_0$ , to an extent that increases monotonically with the strength of the applied field. Along the direction  $\zeta_3$ , in contrast, a compression which arises in the material for lower applied fields turns into an expansion for the stronger values of the magnetic field.

To explain the behavior predicted in Table 1 we focus attention on a generic pair of particles, as depicted in Fig. 3. The location of  $\Omega_2$  with respect to  $\Omega_1$  in the unperturbed state is given by the vector  $\mathbf{r}$ ; all other particles in  $V$  are assumed far away from  $\Omega_1$  and  $\Omega_2$ . To visualize some of the interactions that take place in the composite we focus attention on a pair of particles with  $r = |\mathbf{r}| = 2.5a$  and various values of the angle between  $\mathbf{r}$  and  $\mathbf{H}_0$ .

To begin our discussion we consider the first experiment in Table 1, where the applied magnetic field is very strong and the magnetization of all particles in  $V$  is nearly saturated. For small values of  $\theta$ , the force balance calculation of Section 4.3 tells us that  $\Omega_1$  and  $\Omega_2$  approach each other: the displacement of  $\Omega_1$  and  $\Omega_2$  essentially lies along the  $\zeta_3$ , and it contributes to the compression of  $V$  in this direction. For an angle  $\theta \in (0, \pi/4] \cup [3\pi/4, \pi)$ , in turn, the displacement of  $\Omega_1$  and  $\Omega_2$  is as shown in Fig. 4: the particles approach each other in all directions  $\zeta_i$ ,  $i = 1, 2, 3$ , and they thus contribute to the overall compression of  $V$ . For an angle  $\theta \in (\pi/4, 2\pi/5] \cup [3\pi/5, 3\pi/4)$  the displacement of the particles is as shown in Fig. 5: the pair  $\{\Omega_1, \Omega_2\}$  still moves towards alignment with  $\mathbf{H}_0$ , but this displacement causes the particles to approach each other in directions  $\zeta_1$  and  $\zeta_2$  and to move away from each other in the direction  $\zeta_3$ . For  $\theta \in (2\pi/5, 3\pi/5)$ , finally, the displacement of the particles is as depicted in Fig. 6: the particles move away from each other; the displacement along  $\zeta_3$  in this case is much smaller than the displacement in the other directions and it vanishes at  $\theta = \pi/2$ .

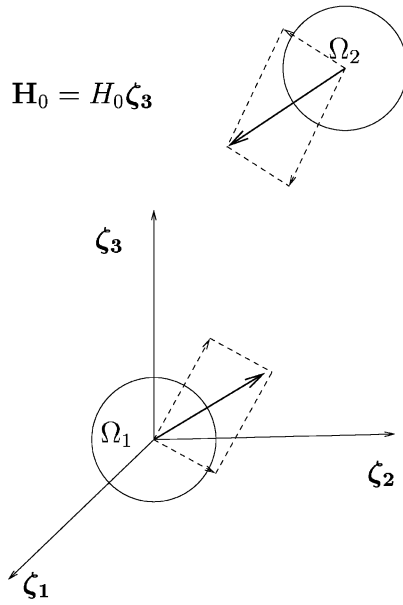


Fig. 4. The particles approach each other along all directions  $\zeta_i$ ,  $i = 1, 2, 3$ .

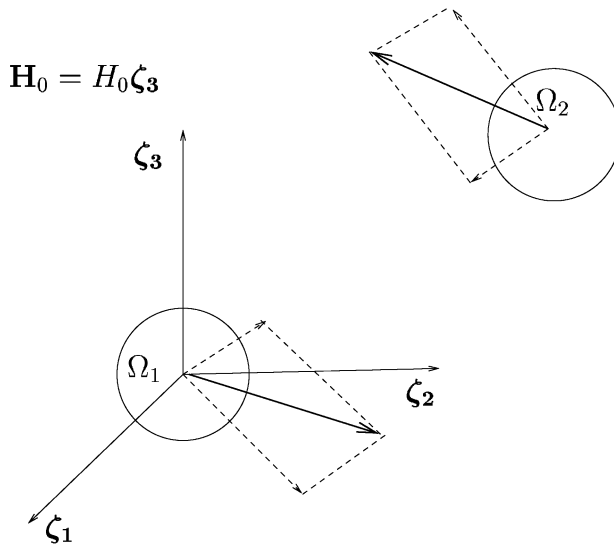


Fig. 5. The particles approach each other along directions  $\zeta_1$  and  $\zeta_2$  and they withdraw from each other along  $\zeta_3$ .

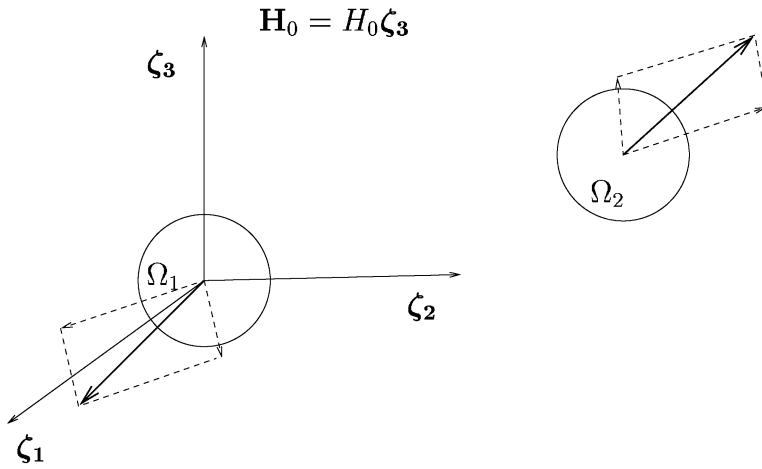


Fig. 6. Particles reject each other.

Table 2  
Self deformation for various shear moduli  $\mu$

$\mu \times 10^5 \text{ dyn/cm}^2$	$\epsilon$	$\tau$	$\epsilon_{11}^0 = \epsilon_{22}^0$	$\epsilon_{33}^0$
7.892	$-4.483 \times 10^{-2}$	$-2.338 \times 10^{-3}$	$-4.716 \times 10^{-2}$	$-4.015 \times 10^{-2}$
8.272	$-3.855 \times 10^{-2}$	$-1.057 \times 10^{-3}$	$-3.961 \times 10^{-2}$	$-3.644 \times 10^{-2}$
9.120	$-3.150 \times 10^{-2}$	$-3.209 \times 10^{-3}$	$-3.471 \times 10^{-2}$	$-2.509 \times 10^{-2}$
11.910	$-2.488 \times 10^{-2}$	$-1.067 \times 10^{-4}$	$-2.499 \times 10^{-2}$	$-2.467 \times 10^{-2}$
16.210	$-9.144 \times 10^{-3}$	$-4.746 \times 10^{-3}$	$-1.389 \times 10^{-2}$	$3.487 \times 10^{-4}$
32.0	$-2.659 \times 10^{-3}$	$-3.936 \times 10^{-3}$	$-6.595 \times 10^{-3}$	$5.213 \times 10^{-3}$

Clearly, the overall deformation of  $V$  is due to the displacement of all particles in the composite. We see that, for the first experiment in Table 1, the dominant displacement of particles along the directions  $\zeta_1$  and  $\zeta_2$  leads to an overall compression of the material in these directions; along  $\zeta_3$ , in contrast, the material is slightly dilated—due to the counter effect of pairs of particles such as those shown in Fig. 5.

For the subsequent experiments in Table 1 we see that, as  $H_0$  decreases, so do the displacements of the particles in  $V$ . The overall compression of the material decreases in directions orthogonal to  $\mathbf{H}_0$ , but it actually increases in direction of  $\mathbf{H}_0$ —since the displacement of particles such as those in Fig. 5 is smaller for weaker  $\mathbf{H}_0$ . Finally, as  $\mathbf{H}_0$  decreases even further, the self-deformation of  $V$  approaches zero in all directions, as expected.

The experiments of Table 2 test the effect of the strength of the matrix on the overall magneto-elastic properties of the composite. In this study we thus fix  $H_0 = 8.595 \times 10^3 \text{ Oe}$  and  $\nu = 0.4$  and we vary the shear modulus  $\mu$ . Naturally, for softer materials the overall deformation increases—since, for softer materials, the magnetic forces are better able to create large displacements within the matrix.

Table 3  
Self deformation for various Poisson ratios  $\nu$

$\nu$	$\varepsilon$	$\tau$	$\varepsilon_{11}^0 = \varepsilon_{22}^0$	$\varepsilon_{33}^0$
0.3	$-1.041 \times 10^{-1}$	$5.553 \times 10^{-3}$	$-1.096 \times 10^{-1}$	$-9.301 \times 10^{-2}$
0.35	$-5.245 \times 10^{-2}$	$-5.079 \times 10^{-3}$	$-5.753 \times 10^{-2}$	$-4.229 \times 10^{-2}$
0.4	$-3.151 \times 10^{-2}$	$-3.203 \times 10^{-3}$	$-3.471 \times 10^{-2}$	$-2.510 \times 10^{-2}$
0.45	$-1.418 \times 10^{-2}$	$-4.802 \times 10^{-3}$	$-1.899 \times 10^{-2}$	$-4.576 \times 10^{-3}$

In Table 3, finally, we fix  $H_0 = 8.595 \times 10^3$  Oe and  $\mu = 9.12 \times 10^5$  dyn/cm<sup>2</sup> and we compute the deformation in the composite for various values of the Poisson ratio  $\nu$ . We see that smaller values of  $\nu$  gives rise to larger overall deformations of  $V$ .

6.3. Average strain for given surface tractions

In this section, we calculate the overall strain in the composite for given surface tractions on  $\partial V$ . We do this for an elastic matrix with the Poisson ratio  $\nu = 0.4$  and shear modulus  $\mu = 9.12 \times 10^5$  dyn/cm<sup>2</sup>, containing a volume fraction  $\Phi = 0.2$  of ferromagnetic particles. The effective bulk modulus  $K^\star = (3\lambda^\star + 2\mu^\star)/3$  and shear modulus  $\mu^\star$  are calculated to order  $O(\Phi^2)$  by Chen and Acrivos (1978b); in this case their calculations give  $3/2\mu K^\star = 9.2252$  and  $\mu^\star/\mu = 1.6007$ .

Let us first assume a surface traction of the form  $S_i = 0.3\mu n_3 \delta_{i3}$ . Without an applied magnetic field the deformation of  $V$  equals

$$\tilde{\varepsilon}_{ij}^0 = -2.582 \times 10^{-2}(\delta_{i1}\delta_{j1} + \delta_{i2}\delta_{j2}) + 6.789 \times 10^{-2}\delta_{i3}\delta_{j3}, \tag{119}$$

corresponding to  $\tilde{\varepsilon} = 5.420 \times 10^{-3}$  and  $\tilde{\tau} = -3.12410^{-2}$ . For an applied field  $H_0 = 7.412 \times 10^3$  Oe, in contrast, the actual deformation of  $V$  equals

$$\varepsilon_{ij}^0 = -5.820 \times 10^{-2}(\delta_{i1}\delta_{j1} + \delta_{i2}\delta_{j2}) + 3.402 \times 10^{-2}\delta_{i3}\delta_{j3}$$

— which gives  $\varepsilon = -2.746 \times 10^{-2}$  and  $\tau = -3.074 \times 10^{-2}$ . Hence, magnetic interactions in  $V$  cause a significant difference in the response of the material to surface tractions. In the directions  $\zeta_1$  and  $\zeta_2$ , for example, the magnetic interactions give rise to a contraction that is twice as large as that observed in the absence of applied magnetic fields.

Next, take  $S_i = -0.3\mu n_3 \delta_{i3}$ . In the absence of a magnetic field, the average strain would be the negative of  $\tilde{\varepsilon}_{ij}^0$  in Eq. (119). For  $H_0 = 7.412 \times 10^3$  Oe, the actual deformation is

$$\varepsilon_{ij}^0 = 7.831 \times 10^{-3}(\delta_{i1}\delta_{j1} + \delta_{i2}\delta_{j2}) - 0.141\delta_{i3}\delta_{j3},$$

which gives  $\varepsilon = -4.172 \times 10^{-2}$  and  $\tau = 4.955 \times 10^{-2}$ . Hence, magnetic interactions in  $V$  oppose the applied forces in directions  $\zeta_1$  and  $\zeta_2$  and cause the material to dilate by a lesser amount than that at  $\mathbf{H}_0 = \mathbf{0}$ . Furthermore, the applied force in direction  $\zeta_3$  is favored by the magnetic interactions in  $V$  and the result is a much larger compression than at zero magnetic field.

Next, take  $S_i = 0.3\mu(n_1\delta_{i1} + n_2\delta_{i2})$ . The deformation in the absence of a magnetic field is

$$\tilde{\varepsilon}_{ij}^0 = 4.208 \times 10^{-2}(\delta_{i1}\delta_{j1} + \delta_{i2}\delta_{j2}) - 5.163 \times 10^{-2}\delta_{i3}\delta_{j3}, \quad (120)$$

so that  $\tilde{\varepsilon} = 1.084 \times 10^{-2}$  and  $\tilde{\tau} = 3.124 \times 10^{-2}$ . In the presence of our magnetic field  $H_0 = 7.412 \times 10^3$  Oe, instead, the actual deformation of  $V$  is

$$\varepsilon_{ij}^0 = 2.120 \times 10^{-2}(\delta_{i1}\delta_{j1} - \delta_{i2}\delta_{j2}) - 0.1094\delta_{i3}\delta_{j3}$$

—or  $\varepsilon = -2.181 \times 10^{-2}$  and  $\tau = 4.381 \times 10^{-2}$ . As in the previous experiments, the magnetic interactions in  $V$  oppose the applied forces that tend to elongate the material in the direction of the applied field, and they contribute towards compressions in the orthogonal directions. The result is a smaller dilation in directions  $\zeta_1$  and  $\zeta_2$  and a larger compression in the direction  $\zeta_3$ .

Finally, for  $S_i = -0.3\mu(n_1\delta_{i1} + n_2\delta_{i2})$ , the overall strain under zero applied magnetic field equals the negative of  $\tilde{\varepsilon}_{ij}^0$  in Eq. (120). Under our applied magnetic field we have, instead

$$\varepsilon_{ij}^0 = -8.435 \times 10^{-2}(\delta_{i1}\delta_{j1} + \delta_{i2}\delta_{j2}) + 2.852 \times 10^{-2}\delta_{i3}\delta_{j3}$$

—or  $\varepsilon = -4.673 \times 10^{-2}$  and  $\tau = -3.763 \times 10^{-2}$ .

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### Appendix A. Solution of elementary problems (69)

The method of solution of the elementary problems (69) is similar to that of Chen and Acrivos (1978a). Given two particles  $\Omega_1$  and  $\Omega_2$  (which, in the unperturbed state, lie at a distance  $r$  from each other). We introduce the coordinate systems  $(x_i, y_i, z_i)$  which are concentric with the centers of  $\Omega_i$ ,  $i = 1, 2$ , respectively, where  $z_i$  are taken along the axis of the centers of the particles, as shown in Fig. 7. Clearly, these systems of coordinates are related by

$$x_1 = x_2, \quad y_1 = y_2, \quad z_1 = z_2 + r.$$

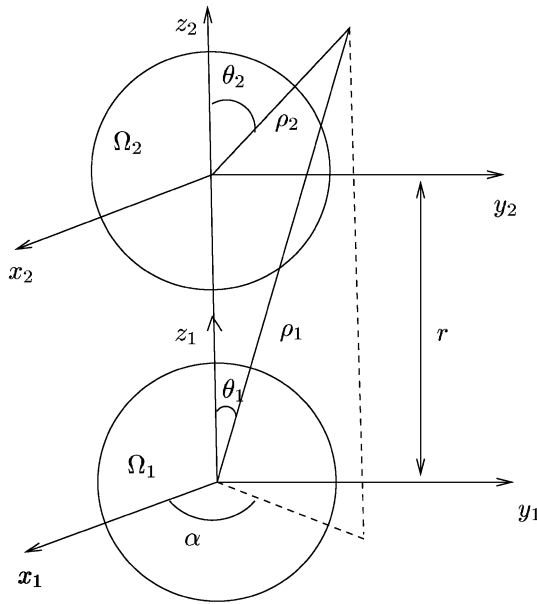


Fig. 7. Coordinates for the two sphere system.

Furthermore, in spherical coordinates, we have

$$\begin{aligned} x_i &= \rho_i \sin \theta_i \cos \alpha, \\ y_i &= \rho_i \sin \theta_i \sin \alpha, \\ z_i &= \rho_i \cos \theta_i, \quad i = 1, 2. \end{aligned}$$

In what follows we provide a detailed calculation for the solution  $\chi_3$ —which satisfies

$$\begin{aligned} \nabla[\nabla \cdot \chi_3(\mathbf{x})] + (1 - 2\nu)\nabla^2 \chi_3(\mathbf{x}) &= 0 \quad \text{in } \mathbb{R}^3 \setminus \{\Omega_1 \cup \Omega_2\}, \\ \chi_3(\mathbf{x}) &= -\frac{a}{2} \mathbf{e}_3 \quad \text{for } \mathbf{x} \in \partial\Omega_1, \\ \chi_3(\mathbf{x}) &= \frac{a}{2} \mathbf{e}_3 \quad \text{for } \mathbf{x} \in \partial\Omega_2. \end{aligned} \tag{A.1}$$

The calculation of the other eleven elementary displacements in Eq. (69) follows similarly.

The solution  $\chi_3$  is given by

$$2\mu\chi_3(\mathbf{x}) = \sum_{i=1}^2 \{ \nabla[\tau^{(i)}(\mathbf{x}) + z_i \tau_3^{(i)}(\mathbf{x})] - 4(1 - \nu)\tau_3^{(i)}(\mathbf{x})\mathbf{e}_3 \}, \tag{A.2}$$

where, using unknown coefficients  $A_m^{(i)}$  and  $B_m^{(i)}$  and Legendre functions  $P_m(\cdot)$  (Abramovitz and Stegun, 1972), we have set

$$\begin{aligned} \tau^{(i)}(\mathbf{x}) &= \sum_{m=0}^{\infty} A_m^{(i)} \frac{a^{m+3}}{\rho_i^{m+1}} P_m(\cos \theta_i) \text{ and} \\ \tau_3^{(i)}(\mathbf{x}) &= \sum_{m=0}^{\infty} B_m^{(i)} \frac{a^{m+2}}{\rho_i^{m+1}} P_m(\cos \theta_i), \quad i = 0, 1. \end{aligned} \tag{A.3}$$

To evaluate the displacement in the spherical system of coordinates  $(\rho_1, \theta_1, \alpha)$  we use the addition theorem (Hobson, 1931)

$$\frac{P_m(\cos \theta_2)}{\rho_2^{m+1}} = (-1)^m \sum_{s=0}^{\infty} \frac{(s+m)!}{s!m!} \frac{\rho_1^s}{r^{s+m+1}} P_s(\cos \theta_1) \tag{A.4}$$

and we obtain

$$\begin{aligned} \chi_3 \cdot \hat{\rho}_1 &= \frac{1}{2\mu} \sum_{m=0}^{\infty} \left\{ -A_m^{(1)} (m+1) \frac{a^{m+3}}{\rho_1^{m+2}} P_m(\cos \theta_1) - B_m^{(1)} \frac{a^{m+2}}{\rho_1^{m+1}} \frac{m+4-4\nu}{2m+1} \right. \\ &\quad \times [(m+1)P_{m+1}(\cos \theta_1) + mP_{m-1}(\cos \theta_1)] + (-1)^m a^{m+2} \left( \frac{a}{r} A_m^{(1)} - B_m^{(1)} \right) \\ &\quad \times \sum_{s=0}^{\infty} \frac{(s+m)!}{s!m!} \frac{s\rho_1^{s-1}}{r^{s+m}} P_s(\cos \theta_1) + (-1)^m a^{m+2} B_m^{(1)} \sum_{s=0}^{\infty} \frac{(s+m)!}{s!m!} \\ &\quad \times \left. \frac{m-3+4\nu}{2s+1} \frac{\rho_1^s}{r^{m+s+1}} [(s+1)P_{s+1}(\cos \theta_1) + sP_{s-1}(\cos \theta_1)] \right\}, \\ \chi_3 \cdot \hat{\theta}_1 &= \frac{1}{2\mu} \sum_{m=0}^{\infty} \left\{ A_m^{(1)} \frac{a^{m+3}}{\rho_1^{m+2}} P_m^1(\cos \theta_1) + B_m^{(1)} \frac{a^{m+2}}{\rho_1^{m+1}} \left[ \frac{m-3+4\nu}{2m+1} P_{m+1}^1(\cos \theta_1) \right. \right. \\ &\quad \left. \left. + \frac{m+4-4\nu}{2m+1} P_{m-1}^1(\cos \theta_1) \right] + (-1)^m a^{m+2} \left( \frac{a}{r} A_m^{(1)} - B_m^{(1)} \right) \sum_{s=0}^{\infty} \frac{(s+m)!}{s!m!} \right. \\ &\quad \times \frac{\rho_1^{s-1}}{r^{s+m}} P_s^1(\cos \theta_1) + (-1)^m a^{m+2} B_m^{(1)} \sum_{s=0}^{\infty} \frac{(s+m)!}{s!m!} \frac{\rho_1^s}{r^{s+m+1}} \\ &\quad \times \left. \left[ \frac{s-3+4\nu}{2s+1} P_{s+1}^1(\cos \theta_1) + \frac{s+4-4\nu}{2s+1} P_{s-1}^1(\cos \theta_1) \right] \right\}, \\ \chi_3 \cdot \hat{\alpha} &= 0. \end{aligned}$$

Similarly, we calculate the displacement in the spherical system of coordinates  $(\rho_2, \theta_2, \alpha)$ . And, finally, imposing the boundary conditions

$$\chi_3 = -\frac{a}{2} [P_1(\cos \theta_1) \hat{\rho}_1 + P_1^1(\cos \theta_1) \hat{\theta}_1] \quad \text{at } \partial\Omega_1,$$

$$\chi_3 = \frac{a}{2} [P_1(\cos \theta_2) \hat{\rho}_2 + P_1^1(\cos \theta_2) \hat{\theta}_2] \quad \text{at } \partial\Omega_2$$

leads to an infinite system of equations which can be used to determine the coefficients  $A_m^{(i)}$  and  $B_m^{(i)}$ . Due to the symmetries in the problem, we have

$$\begin{aligned}
 A_m^{(2)} &= (-1)^m A_m^{(1)}, \\
 B_m^{(2)} &= (-1)^{m+1} B_m^{(1)}, \quad m = 0 \dots \infty
 \end{aligned}
 \tag{A.5}$$

and it therefore suffices to calculate the coefficients for particle  $\Omega_1$  — which we rename as

$$\begin{aligned}
 A_m^{(1)} &= 2\mu A_m^{\zeta_3}, \\
 B_m^{(1)} &= 2\mu B_m^{\zeta_3}, \quad m = 0 \dots \infty.
 \end{aligned}
 \tag{A.6}$$

Notation (A.6) allows us to distinguish between the coefficients needed for the solution of each one of the elementary problems in Eq. (69). The coefficients in Eq. (A.6) can be obtained rather inexpensively—through use of series expansions in powers of  $a/r$ . Indeed, letting

$$\begin{aligned}
 A_m^{\zeta_3} &= \frac{1}{4(6\nu - 5)} \delta_{m1} + \sum_{p=0}^{\infty} \alpha_{mp} \left(\frac{a}{r}\right)^{m+p}, \\
 B_m^{\zeta_3} &= -\frac{3}{4(6\nu - 5)} \delta_{m0} + \sum_{p=0}^{\infty} \beta_{mp} \left(\frac{a}{r}\right)^{m+p+1}, \quad m = 0 \dots \infty
 \end{aligned}
 \tag{A.7}$$

one easily finds recursively the expression for  $\alpha_{mp}$  and  $\beta_{mp}$  for  $m = 0 \dots \mathcal{M}$  and  $p = 0 \dots \mathcal{P}$ —where the truncation parameters  $\mathcal{M}$  and  $\mathcal{P}$  are to be determined from convergence tests of expansions (A.7) and (A.3).

In our calculations of the net force (70) and torque (72) we need expressions for the tractions on the surface of  $\Omega_1$ ; algebraic manipulations yield the expressions

$$\begin{aligned}
 &\frac{\sigma_{\rho\rho}^{\zeta_3}(a, \theta_1, \alpha)}{2\mu} \\
 &= \sum_{m=0}^{\infty} \left\{ \frac{(m+2)!}{m!} A_m^{\zeta_3} P_m(\cos \theta_1) + \frac{m+1}{2m+1} B_m^{\zeta_3} [(m+4-4\nu)mP_{m-1}(\cos \theta_1) \right. \\
 &\quad \left. + (m^2 + 5m + 4 - 2\nu)P_{m+1}(\cos \theta_1)] + \left(\frac{a}{r} A_m^{\zeta_3} + B_m^{\zeta_3}\right) \sum_{s=0}^{\infty} \frac{(m+s)!s(s-1)}{s!m!} \right. \\
 &\quad \times \left(\frac{a}{r}\right)^{m+s} P_s(\cos \theta_1) - B_m^{\zeta_3} \sum_{s=0}^{\infty} \frac{(m+s)!}{s!m!} \left(\frac{a}{r}\right)^{m+s+1} \left[ \frac{s(s^2 - 3s - 2\nu)}{2s+1} \right. \\
 &\quad \left. \left. \times P_{s-1}(\cos \theta_1) + \frac{s(s+1)(s-3+4\nu)}{2s+1} P_{s+1}(\cos \theta_1) \right] \right\},
 \end{aligned}$$

$$\begin{aligned} \frac{\sigma_{\rho\theta}^{\chi_3}(a, \theta_1, \alpha)}{2\mu} &= \sum_{m=0}^{\infty} \left\{ -(m+2)A_m^{\chi_3} P_m^1(\cos \theta_1) \right. \\ &\quad \left. - B_m^{\chi_3} \left[ \frac{(m+1)(m+4-4\nu)}{2m+1} P_{m-1}^1(\cos \theta_1) + \frac{m^2+2m-1+2\nu}{2m+1} P_{m+1}^1(\cos \theta_1) \right] \right. \\ &\quad \left. + \left( \frac{a}{r} A_m^{\chi_3} + B_m^{\chi_3} \right) \sum_{s=0}^{\infty} \frac{(m+s)!(s-1)}{s!m!} \left( \frac{a}{r} \right)^{m+s} P_s^1(\cos \theta_1) \right. \\ &\quad \left. - B_m^{\chi_3} \sum_{s=0}^{\infty} \frac{(m+s)!}{s!m!} \left( \frac{a}{r} \right)^{m+s+1} \left[ \frac{s^2-2+2\nu}{2s+1} P_{s-1}^1(\cos \theta_1) \right. \right. \\ &\quad \left. \left. + \frac{s(s-3+4\nu)}{2s+1} P_{s+1}^1(\cos \theta_1) \right] \right\}, \end{aligned}$$

$$\sigma_{\rho z}^{\chi_3}(a, \theta_1, \alpha) = 0.$$

Thus, the net force  $\mathbf{F}^{\chi_3(1)}$ , acting on  $\Omega_1$  and due to the displacement  $\chi_3$  is given by

$$\begin{aligned} \mathbf{F}^{\chi_3(1)} \cdot \mathbf{e}_1 &= a^2 \int_0^\pi \int_0^{2\pi} [\sigma_{\rho\rho}^{\chi_3} \sin \theta_1 \cos \alpha \\ &\quad + \sigma_{\rho\theta}^{\chi_3} \cos \theta_1 \cos \alpha - \sigma_{\rho z}^{\chi_3} \sin \alpha] \sin \theta_1 d\theta_1 d\alpha = 0, \\ \mathbf{F}^{\chi_3(1)} \cdot \mathbf{e}_2 &= a^2 \int_0^\pi \int_0^{2\pi} [\sigma_{\rho\rho}^{\chi_3} \sin \theta_1 \sin \alpha \\ &\quad + \sigma_{\rho\theta}^{\chi_3} \cos \theta_1 \sin \alpha + \sigma_{\rho z}^{\chi_3} \cos \alpha] \sin \theta_1 d\theta_1 d\alpha = 0, \\ \mathbf{F}^{\chi_3(1)} \cdot \mathbf{e}_3 &= a^2 \int_0^\pi \int_0^{2\pi} [\sigma_{\rho\rho}^{\chi_3} \cos \theta_1 - \sigma_{\rho\theta}^{\chi_3} \sin \theta_1] \sin \theta_1 d\theta_1 d\alpha \\ &= 16\pi\mu a^2(1-\nu)B_0^{\chi_3}. \end{aligned} \tag{A.8}$$

Furthermore, the torque acting on  $\Omega_1$  is  $\mathcal{T}_i^{\chi_3(1)} = 0$ , for  $i = 1, 2, 3$ . Naturally, the forces on  $\Omega_2$  are given by

$$F_i^{\chi_3(2)} = -F_i^{\chi_3(1)}, \quad \mathcal{T}_i^{\chi_3(2)} = 0, \quad i = 1, 2, 3. \tag{A.9}$$

**Appendix B. Proof that, at saturation of magnetization, the magnetic energy  $\mathcal{W}_{1,2}^{\text{mag}}$  given by Eq. (77) is minimized by  $\mathbf{M}^{(1)} = \mathbf{M}^{(2)}$**

Suppose that the applied magnetic field  $\mathbf{H}_0$  is sufficiently strong such that the magnetization of the particles  $\Omega_1$  and  $\Omega_2$  has reached saturation. Then,  $|\mathbf{M}^{(p)}| = M^{\text{sat}}$ , where  $M^{\text{sat}}$  is the saturation magnetization of the ferromagnetic particles. With the notation

$\rho = a/|\mathbf{r}^{\text{eq}}|$ , we define the functional

$$\mathcal{W} = -\frac{4\pi a^3}{3}(\mathbf{M}^{(2)} + \mathbf{M}^{(1)}) \cdot \mathbf{H}_0 + \frac{16\pi^2 a^3}{9} \times \rho^3 [\mathbf{M}^{(2)} \cdot \mathbf{M}^{(1)} - 3(\mathbf{M}^{(2)} \cdot \mathbf{e}'_3)(\mathbf{M}^{(1)} \cdot \mathbf{e}'_3)], \tag{B.1}$$

whose minimizers—over all orientations of  $\mathbf{M}^{(1)}$  and  $\mathbf{M}^{(2)}$ —are the magnetizations we seek. Note that the functional in Eq. (B.1) differs from the magnetic energy  $\mathcal{W}_{1,2}^{\text{mag}}$  in Eq. (77) by the known (constant) term  $8\pi^2/9a^3(|\mathbf{M}^{(2)}|^2 + |\mathbf{M}^{(1)}|^2) = 16\pi^2/9a^3(M^{\text{sat}})^2$ .

Introducing the angles  $\theta_i \in [0, \pi]$  between  $\mathbf{M}^{(i)}$  and  $\mathbf{e}'_3$ , where  $i = 1, 2$ , the magnetizations can be expressed in the form

$$\mathbf{M}^{(i)} = M^{\text{sat}}(\sin \theta_i \cos \phi_i \mathbf{e}'_1 + \sin \theta_i \sin \phi_i \mathbf{e}'_2 + \cos \theta_i \mathbf{e}'_3), \quad i = 1, 2, \tag{B.2}$$

where  $\phi_i \in [0, 2\pi]$ . Similarly, we may write

$$\mathbf{H}_0 = H_0(\sin \psi \cos \lambda \mathbf{e}'_1 + \sin \psi \sin \lambda \mathbf{e}'_2 + \cos \psi \mathbf{e}'_3). \tag{B.3}$$

With these notations we now may establish the following result.

**Lemma 1.** *The minimizers of functional (B.1) satisfy*

$$\mathbf{M}^{(1)} = \mathbf{M}^{(2)},$$

and, hence, with  $\theta_1 = \theta_2 = \theta$  and  $\phi_1 = \phi_2 = \phi$  in Eq. (B.2), functional (B.1) at saturation becomes

$$\mathcal{W} = -\frac{8\pi a^3}{3} M^{\text{sat}} H_0 [\sin \theta \sin \psi \cos(\lambda - \phi) + \cos \theta \cos \psi] + \frac{16\pi^2 a^3 \rho^3}{9} (M^{\text{sat}})^2 (1 - 3 \cos^2 \theta). \tag{B.4}$$

**Proof of Lemma 1.** By contradiction, assume

$$\mathbf{M}^{(1)} \neq \mathbf{M}^{(2)}. \tag{B.5}$$

The first order optimality conditions (Gill et al., 1989) that the magnetizations  $\mathbf{M}^{(1)}$  and  $\mathbf{M}^{(2)}$  satisfy imply that

$$\mathbf{H}_0 \cdot (\mathbf{p}^{(1)} + \mathbf{p}^{(2)}) = \frac{4\pi \rho^3}{3} [\mathbf{M}^{(1)} \cdot \mathbf{p}^{(2)} + \mathbf{M}^{(2)} \cdot \mathbf{p}^{(1)} - 3(\mathbf{e}'_3 \cdot \mathbf{p}^{(1)})(\mathbf{e}'_3 \cdot \mathbf{M}^{(2)}) - 3(\mathbf{e}'_3 \cdot \mathbf{p}^{(2)})(\mathbf{e}'_3 \cdot \mathbf{M}^{(1)})], \tag{B.6}$$

for all vectors  $\mathbf{p}^{(i)}$  such that  $\mathbf{p}^{(i)} \cdot \mathbf{M}^{(i)} = 0$ ,  $i = 1, 2$ .

We now define a unit vector  $\mathbf{q}$  orthogonal to the plane (or line)  $\text{span}\{\mathbf{M}^{(1)}, \mathbf{M}^{(2)}\}$ . Using  $\mathbf{p}^{(1)} = \mathbf{p}^{(2)} = \mathbf{q}$  in Eq. (B.6) we obtain

$$\mathbf{H}_0 \cdot \mathbf{q} = -4\pi \rho^3 (\mathbf{q} \cdot \mathbf{e}'_3)(\mathbf{M}^{(1)} \cdot \mathbf{e}'_3 + \mathbf{M}^{(2)} \cdot \mathbf{e}'_3); \tag{B.7}$$

letting  $\mathbf{p}^{(1)} = -\mathbf{p}^{(2)} = \mathbf{q}$ , in turn, we obtain

$$(\mathbf{q} \cdot \mathbf{e}'_3)(\mathbf{M}^{(1)} \cdot \mathbf{e}'_3 - \mathbf{M}^{(2)} \cdot \mathbf{e}'_3) = 0. \tag{B.8}$$

In view of Eq. (B.8) we see we must analyze two different cases.

Case 1.  $\mathbf{M}^{(1)} \cdot \mathbf{e}'_3 = \mathbf{M}^{(2)} \cdot \mathbf{e}'_3$  or, equivalently,  $\theta_1 = \theta_2 = \theta$ . In this case, using  $\mathbf{p}^{(i)} = \sin \phi_i \mathbf{e}'_1 - \cos \phi_i \mathbf{e}'_2$ ,  $i = 1, 2$  in Eq. (B.6) we obtain

$$\sin \psi \sin \left( \lambda - \frac{\phi_1 + \phi_2}{2} \right) \cos \left( \frac{\phi_2 - \phi_1}{2} \right) = 0. \tag{B.9}$$

Next, using

$$\mathbf{p}^{(1)} = -\cos \theta \cos \phi_1 \mathbf{e}'_1 - \cos \theta \sin \phi_1 \mathbf{e}'_2 + \sin \theta \mathbf{e}'_3$$

and

$$\mathbf{p}^{(2)} = \cos \theta \cos \phi_2 \mathbf{e}'_1 + \cos \theta \sin \phi_2 \mathbf{e}'_2 - \sin \theta \mathbf{e}'_3.$$

Eq. (B.6) gives

$$\sin \psi \sin \left( \lambda - \frac{\phi_1 + \phi_2}{2} \right) \cos \theta = 0. \tag{B.10}$$

Finally, taking  $\mathbf{p}^{(1)} = -\sin \phi_1 \mathbf{e}'_1 + \cos \phi_1 \mathbf{e}'_2$  and  $\mathbf{p}^{(2)} = \sin \phi_2 \mathbf{e}'_1 - \cos \phi_2 \mathbf{e}'_2$  we obtain, from Eq. (B.6),

$$\frac{8\pi\rho^3}{3} M^{\text{sat}} \sin \theta \cos \left( \frac{\phi_2 - \phi_1}{2} \right) = H_0 \sin \psi \cos \left( \lambda - \frac{\phi_1 + \phi_2}{2} \right). \tag{B.11}$$

Suppose that  $\sin \psi \sin(\lambda - (\phi_1 + \phi_2)/2) \neq 0$  such that Eqs. (B.9) and (B.10) give  $\cos \theta = \cos(\phi_2 - \phi_1/2) = 0$  or, equivalently,  $\mathbf{M}^{(1)} = -\mathbf{M}^{(2)}$ . The energy given by this choice of magnetizations is

$$\mathcal{W} = -\frac{16\pi^2 a^3 \rho^3}{9} (M^{\text{sat}})^2. \tag{B.12}$$

This cannot be a global minimum of Eq. (B.1). Indeed, take in Eq. (B.4) an angle  $\phi$  equal to  $\lambda$  and choose  $\theta$  either 0 or  $\pi$  such that  $\cos(\psi - \theta) = |\cos(\psi - \theta)|$ . The result in Eq. (B.4) is a smaller energy than that of Eq. (B.12). Thus, we must have

$$\sin \psi \sin \left( \lambda - \frac{\phi_1 + \phi_2}{2} \right) = 0. \tag{B.13}$$

If  $\sin \psi = 0$ , the applied magnetic field is along the axis  $\mathbf{e}'_3$  and Eq. (B.11) gives

$$\sin \theta \cos \left( \frac{\phi_2 - \phi_1}{2} \right) = 0.$$

Assumption (B.5) holds so,  $\sin \theta \neq 0$ . Thus, the angles  $\phi_1$  and  $\phi_2$  are related by

$$\phi_2 = \phi_1 + (2k + 1)\pi, \tag{B.14}$$

where  $k$  is an integer. The energy in this case is

$$\mathcal{W} = -\frac{8\pi a^3}{3} M^{\text{sat}} H_0 \cos \theta - \frac{16\pi^2 a^3 \rho^3}{9} (M^{\text{sat}})^2 (1 + \cos^2 \theta), \quad (\text{B.15})$$

where  $\cos \theta \neq 1$ . However, if in Eq. (A.4), we let  $\theta = 0$ , we obtain a smaller energy than Eq. (B.15). Hence, the only alternative left to explore is

$$\sin\left(\lambda - \frac{\phi_1 + \phi_2}{2}\right) = 0 \quad \text{and} \quad \sin \theta \neq 0. \quad (\text{B.16})$$

Calculations similar to the above show that the energy  $\mathcal{W}$  given in this case is larger than the energy (B.4) obtained with  $\mathbf{M}^{(1)} = \mathbf{M}^{(2)}$ .

*Case 2.* We have  $\mathbf{q} \cdot \mathbf{e}'_3 = 0$  or, equivalently,  $\mathbf{e}'_3 \in \text{span}\{\mathbf{M}^{(1)}, \mathbf{M}^{(2)}\}$ . Here, Eq. (B.7) leads to the conclusion that  $\mathbf{H}_0 \in \text{span}\{\mathbf{M}^{(1)}, \mathbf{M}^{(2)}\}$ , as well. In the plane spanned by the two magnetizations, let  $\mathbf{e}'_3$  and  $\zeta$  be two orthogonal axes. Then, we write

$$\mathbf{M}^{(i)} = M^{\text{sat}} (\sin \theta_i \zeta + \cos \theta_i \mathbf{e}'_3), \quad i = 1, 2, \quad (\text{B.17})$$

$$\mathbf{H}_0 = H_0 (\sin \psi \zeta + \cos \psi \mathbf{e}'_3). \quad (\text{B.18})$$

Let us take in Eq. (B.6),  $\mathbf{p}^{(1)} = -\cos \theta_1 \zeta + \sin \theta_1 \mathbf{e}'_3$  and  $\mathbf{p}^{(2)} = \cos \theta_2 \zeta - \sin \theta_2 \mathbf{e}'_3$ . We obtain

$$\sin\left(\frac{\theta_2 - \theta_1}{2}\right) \left[ \frac{4\pi\rho^3}{3} M^{\text{sat}} \cos\left(\frac{\theta_2 - \theta_1}{2}\right) + H_0 \cos\left(\psi - \frac{\theta_1 + \theta_2}{2}\right) \right] = 0$$

and, since Eq. (B.5) implies  $\sin(\theta_2 - \theta_1/2) \neq 0$ ,

$$H_0 \cos\left(\psi - \frac{\theta_1 + \theta_2}{2}\right) = -\frac{4\pi\rho^3}{3} M^{\text{sat}} \cos\left(\frac{\theta_2 - \theta_1}{2}\right). \quad (\text{B.19})$$

Furthermore, the energy in Eq. (B.1) becomes

$$\begin{aligned} \mathcal{W} = & -\frac{8\pi a^3 M^{\text{sat}} H_0}{3} \cos\left(\psi - \frac{\theta_1 + \theta_2}{2}\right) \cos\left(\frac{\theta_2 - \theta_1}{2}\right) \\ & + \frac{16\pi^2 a^3 \rho^3 (M^{\text{sat}})^2}{9} [\cos(\theta_2 - \theta_1) - 3 \cos \theta_1 \theta_2]. \end{aligned} \quad (\text{B.20})$$

We note that, due to Eq. (B.19), the first term in the right-hand side of Eq. (B.20) is strictly positive. Since at saturation of the magnetization the applied magnetic field  $H_0$  is large, we cannot obtain a small energy if Eq. (B.19) holds. This can be easily verified by direct calculation and a comparison with the energy in Eq. (B.4), where  $\lambda = \phi$ . Thus, Case 2 cannot give a global minimizer, either. The conclusion is that assumption (B.5) is false and Lemma 1 is proved.

To conclude our analysis we point out that, with a similar method, one can rule out the possibility that just one particle in the pair  $\Omega_1, \Omega_2$  is saturated.

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