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Sound propagation in magneto-rheological suspensions

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Abstract

The propagation of elastic perturbations in magneto-rheological suspensions is studied theoretically and experimentally. Under the application of a magnetic field, these systems acquire a fibrillar fractal structure formed by clusters. In systems in that condition, two low-frequency sound propagation modes have been observed. In both of them, the speed of sound depends on the intensity of the applied field. We discuss the statistical fractal properties of the cluster structure and, on this basis, we calculate the speed of sound for both of the low-frequency modes. This theoretical approach provides a good quantitative agreement with the experimental results.

Magneto-rheological (MR) slurries are composed of micrometric magnetic particles suspended in inert media; glycerine and silicone oil are commonly used. In the presence of a magnetic field, these complex fluids suffer a structural transition which produces an abrupt change in their mechanical properties: change from a viscous fluid to an almost solid body. Of course, in this transition the shear modulus changes by several orders of magnitude. It has been observed that, for low particle concentration (<0.01 in volume fraction) and in the presence of a magnetic field, the particles form a structure which can be well described as chains going from one of the extremes of the observation cell to the other [1]. However, for higher volume fractions, the particles form a much more complex structure. It has been observed that this structure is formed by clusters whose size and shape depend in a complex way on several physical conditions—for instance, magnetic field intensity, particle size and particle shape [2]; nevertheless, the cluster structure exhibits fractal characteristics. We shall use this feature of the cluster structure in describing the elastic properties of these magnetic dispersions. The observation of two sound propagation longitudinal modes in MR slurries has been reported, in the large-wavelength regime [3]. One of these modes has been interpreted as the propagation of sound through channels of almost clear fluid. The other mode, which appears at larger wavelength, has been interpreted as the sound propagation through the structure formed by the particles. In both of these modes the sound speed depends on the intensity of the applied magnetic field. We have prepared soft magnetic iron oxide particles with size ranging from 8 to 10 μm; they were dispersed in silicone oil, and we studied the dynamics of the cluster formation as well as the elastic properties of these MR slurries, under different physical conditions. In this work our statistical characterization of the cluster structure is discussed. We propose a simple effective-media approximation for calculating the speed of propagation of elastic perturbations through these media. This approach is applied to provide a basis for discussion of some results on sound speed reported recently [3].

Clusters formed by the suspended particles under the influence of an applied magnetic field are substructures with fractal properties; however, locally, in the range of a few times the mean size of the particles, they statistically exhibit a crystalline-like structure [4]. In the formation of these substructures two mechanisms are involved: the first one is an ordering mechanism due to the magnetic interaction among the particles and with the external field; and the other one, with a thermal and entropic origin, is a disordering mechanism. The balance between them determines the cluster structure and network, and consequently the rheological response of the system. In order to statistically characterize the geometrical features of the clusters, by using heuristic argumentation and some phenomenological input we have obtained the following cluster size distribution:

$$R(x) = \alpha(\phi)x^{D\lambda} \exp\left(-\frac{D\lambda}{\Sigma}x\right). \tag{1}$$

Here the λ-parameter is given by $\lambda = mB/kT$ (m being the magnetic moment of the particles), B is an effective Weiss-like magnetic field, kT is the thermal energy, D is the measured fractal dimension, ϕ is the particle volume fraction, x describes the cluster longitudinal length and, for a given ϕ, $\alpha(\phi)$ is a normalization constant. It is easy to show that the quantity Σ is the most frequent cluster size. Thus, this is not a fitting parameter, but can be measured, as can the fractal dimension. The functional form of this distribution is typical of those which describe certain physical properties of systems having complex correlations [5]. The iron oxide particles that we dispersed in silicon oil were obtained by coprecipitation of iron (III) nitrate, iron (II) chloride and ammonium oxalate solutions in order to obtain iron oxalate; this was followed by its decomposition at 300 °C. These particles have prismatic shape, and we have detected particles of three of the phases of the iron oxide—namely, magnetite, maghemite and haematite. We have observed, by optical microscopy, that even at relatively low volume fraction and under a weak applied field, cluster formation occurs. In figure 1 we depict the measured frequency cluster size distribution, i.e., the frequency at which clusters with that size were observed. The measurements were performed at room temperature, under a field of 500 G, with a volume fraction of particles $\phi = 0.07$. The continuous line is the distribution obtained from expression (1). Despite the conditions—namely, low volume fraction and low ordering energy—the cluster mean size is of the order of 26 times the particle mean size, σ. This figure shows an illustrative situation of the agreement between the data obtained using equation (1) and the experimental results. We have tested this agreement for a wide range of ϕ- and λ-values and we always observed a similar accord. The inset shows a representative pair of distributions for two values of applied field, 500 and 350 G; it can be observed that the stronger ordering interaction μB promotes a larger mean cluster size. It is worthy of mention that in order to evaluate $R(x)$ for these two cases, we have used expression (1) with the proper value of λ, but with no further changes. From our observations we have concluded that the fractal dimension D has a weak dependence on the magnitude of the applied magnetic field. This is shown in the lower part of the inset.

Now that we have this description of the geometrical cluster features, we shall apply it to provide a basis for discussion of the sound propagation in a MR slurry.

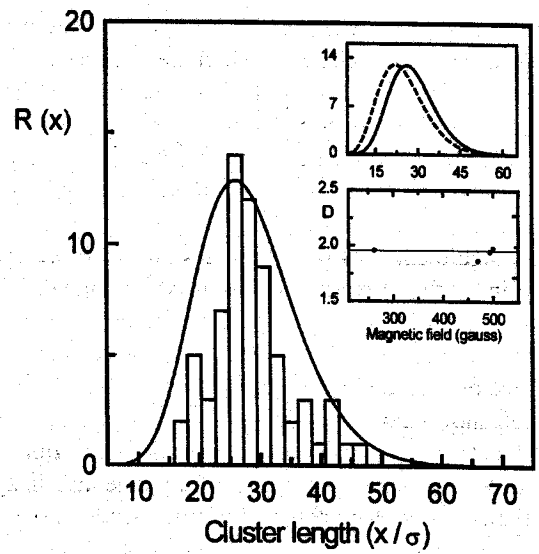

Figure 1. The cluster size distribution $R(x)$ at room temperature, applied field 500 G, $\phi = 0.07$. The cluster length is measured in units of particle mean size, σ. Upper figure in the inset: the cluster size distribution for two values of applied field: (continuous curve) 500 G; (dashed curve) 350 G. Lower figure: the measured fractal dimension, D, for different values of the applied field.

It is well known that in a colloidal suspension, only one longitudinal mode can exist when an elastic excitation propagates in it if the wavelength is much larger than the characteristic size of the suspended particles—although, when the wavelength of the elastic excitation is of the order of the size of the particles, two longitudinal modes have been observed in systems of this kind [6]. Thus, the observation of two distinct modes propagating in the low-frequency regime in a MR slurry composed of iron particles dispersed in glycerine [3] was quite unexpected. The authors' interpretation of this result was that one of these modes corresponds to the propagation of an elastic perturbation through the fluid phase and the other mode propagates through the suspended particles, arranged in perfect chain structures. Furthermore, they found that the second mode is slower than the first one and appears only while the external magnetic field is still being applied. They also found that the speed of propagation of both elastic modes depends on the intensity of the applied magnetic field. Now, we focus our attention on both of these propagation modes and calculate the sound speed by the following procedure. We use our characterization of the cluster fractal properties, as given by distribution (1), and incorporate this in a simple effective-media approximation. This approximation consists in calculating the sound speed in the composite medium v, in terms of an effective density ρ and an effective elastic modulus β, by means of the expressions [7]

$$v = (\beta/\rho)^{1/2} \qquad \rho = \phi\rho_s + (1-\phi)\rho_f \qquad \frac{1}{\beta} = \frac{\phi}{\beta_s} + (1-\phi)\frac{1}{\beta_f}. \tag{2}$$

Here ϕ is the volume fraction of the particles; subscripts s and f stand for solid and fluid respectively. In calculating the sound speed, we assume that in the first mode, the elastic perturbation propagates through channels of almost clear fluid. Provided that the perturbation wavelength is much larger than the characteristic size of the iron particles, one can assume that the effect of the iron spheres in this dilute dispersion is only of inertial nature, i.e., the elastic properties of the fluid will not be affected by the presence of a few non-interacting particles. Then, neglecting dissipation effects, the only average which must be considered in equation (2) is over the density ρ. One can expect this scheme to be more suitable if a strong magnetic field is present. This is so because under that condition, more particles will be stuck to the fibrillar structure, and consequently fewer of them will be in the fluid channels. The second mode has been observed at even larger wavelengths and only appears in the presence of a magnetic field. The fibrillar structure acquired by the suspension upon the application of a magnetic field is the medium in which this mode propagates. Only under very specific conditions of volume fraction and applied magnetic field do the particles form regular chains with cross sections about equal to the average diameter of the particles [1, 8]. Moreover, it has been observed that a more general trend is the formation of clusters which pile up in the cell along the field direction from one extreme to the other [2]. The size and geometry of the clusters are predominantly determined by the morphology and magnetic dipoles of the particles, which, additionally to other physical factors like applied magnetic field and temperature, determine its fractal nature [1]. As we shall show below, the size and shape of the clusters also influence the elastic properties of the MR suspension. In order to calculate the sound speed for the second mode, we consider the following physical picture. We assume that, in agreement with the fractal nature of some of the physical characteristics of these slurries, one may conceive of the system as composed of two coexisting dispersions. One of them is a dilute dispersion of particles with sizes distributed in the range 3–25 μm suspended in the fluid. The first mode propagates in this medium as we discussed above. The other dispersion is formed by the clusters suspended in the fluid. The second mode propagates in this medium. Therefore, in this picture, to calculate the sound speed for the second mode, we may proceed as we did for the first one, provided that the wavelength of the elastic perturbation is much larger than Σ; however, it is necessary to know the effective density and the elastic modulus of the clusters dispersed in the fluid. Using the cluster size distribution (1), the evaluation of the effective density is direct. In a previous work we have studied, by means of this effective-media approximation, a similar situation [9]; in that report we estimated externally the value of ϕ in the cluster channel formed by means of a fitting to the experimental data, whereas here, by using equation (1), the ϕ-value can be easily obtained—although the evaluation of the elastic modulus might require some discussion. The dipolar force between two spheres whose centres are separated by a distance r is given by

$$F = -(3\mu_0/2\pi)(m^2/r^4) \tag{3}$$

where μ_0 is the magnetic permeability. From here, dividing by the cross sectional area of the structure formed by the clusters, the longitudinal stress is obtained. Then, taking the derivative with respect to the longitudinal strain, i.e., $r(\partial/\partial r)$, one obtains an elastic modulus $\beta = 2/3\mu_0 M^2$, where M is the magnetization.

The behaviour of the sound speed as a function of the volume fraction can be seen for both modes in figure 2(a). Our calculation of the first-mode speed is shown by the lower curve; dots show the experimental results. The continuous line was obtained by averaging just the density values, taking into account that for a dilute suspension one can neglect the effects of suspended particles on the elastic properties of the composite medium. The upper pair of curves correspond to the second-mode speed. In this case the scale appears on the right-hand side. The dashed curve is obtained from equation (2), by calculating ρ and β from the corresponding values for the clusters and the fluid. The arrow indicates the value $\phi = 0.32$ obtained by using equation (1). Here we consider the saturation value of the magnetization and an applied magnetic field of 500 G, as was assumed in [3]. The continuous curve is calculated by the same procedure for a more realistic value of the magnetization at this field, namely, $M = 0.97M_s$, where M_s is the saturation value of the magnetization. Figure 2(b) shows our calculation for the second-mode speed as a function of the applied magnetic field

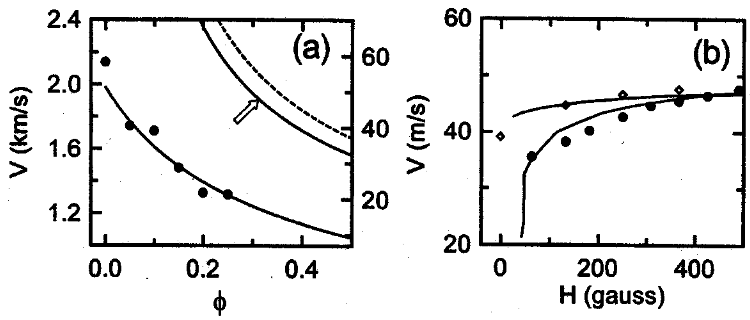

Figure 2. (a) Sound speed as a function of volume fraction ϕ. First mode: lower curve and experimental results (dots). Second mode: upper pair, with the scale on the right. (b) Sound speed versus magnetic field intensity; experimental results for increasing (dots) and decreasing (diamonds) field values.

(continuous curve). Dots show the experimental results for increasing values of magnetic field and the diamonds correspond to decreasing ones, as reported in [3]. For this calculation we have made use of a typical iron magnetization curve. We notice that throughout the interval in which we can compare with the reported measurements, we obtain a good agreement. In [3] for a volume fraction $\phi = 0.25$ and for an applied field of 500 G, by using a model of perfect chains, the authors estimate a sound velocity of $v = 20\,\mathrm{m\,s^{-1}}$. For that condition, our procedure yields a better agreement.

The simple effective-media approximation that we have used here provides a quantitative, accurate description for the sound speed, for both longitudinal modes observed in a MR slurry of spherical iron particles. This approach is based on the statistical description of the fractal properties of the clusters provided by expression (1). We remark that this cluster size distribution does not contain fitting parameters, since the fractal dimension D and the most frequent size Σ can be measured experimentally. The quantity $\alpha(\phi)$, for a given particle concentration, becomes a normalization constant which, per unit volume, approximately satisfies the condition $R(\Sigma)\Sigma^3 = N\sigma^3$, where σ is the particle mean size.

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