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Sound velocity in a magnetic fluid

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Abstract. A simple linearized hydrodynamical theory for magnetic fluids in the presence of a strong external magnetic field is presented. The equations are solved for a sound wave propagating at angle ϕ from the external field direction. The sound velocity is shown to be anisotropic, depending on ϕ. The anisotropy is estimated to be about 10^{-5} for a field of 10^4 G and shows an interesting frequency dependence.

1. Introduction

A magnetic fluid is a colloid of tiny (100 Å) magnetic particles or grains suspended in a carrier fluid such as water (Rosenweig 1966). The magnetization of the fluid varies with the applied magnetic field H, typically reaching a saturation of 10^2–10^3 G for large fields. The grains are kept in suspension by thermal agitation. In addition, to help reduce coagulation, the particles are coated with a dispersing agent such as oleic acid. Nevertheless, because of the magnetic attraction between the grains some coagulation does occur, the extent of which depends on the colloidal number density, the temperature, and the applied magnetic field. At low number densities the grains will exist either singly (monomers) or in pairs (dimers) and it is possible to calculate the relative number of dimers and monomers using statistical mechanics arguments (Jordan 1973). As the concentration of grains increases several of them will combine to form long needles or chains. The chain length increases with concentration and decreases with temperature and is large enough ($\sim$20 μm) to be observed by an optical microscope (Hayes 1975). In the presence of an external magnetic field the chains line up along the direction of the field. It has been suggested (de Gennes and Pincus 1973) that the coagulation can be interpreted in terms of 'liquid' and 'solid' states of the grains separated by definite phase transitions.

The hydrodynamics of a magnetic fluid has been discussed by Jenkins (1972) who included the local magnetization $\boldsymbol{m}(\boldsymbol{r}, t)$ as an extra hydrodynamical variable independent of the usual variables such as pressure, density, temperature and velocity. In the presence of a strong external field, $\boldsymbol{m}$ should be locked nearly parallel to $\boldsymbol{H}$ and deviations from this direction will be small. In this case it is more convenient to separate $\boldsymbol{m}$ into a scalar representing the concentration of the grains, and a unit vector $\boldsymbol{n}$ (the director) which lies along the average orientation of the magnetic easy axis of the grains. We assume that the grains are uniaxial magnetically and the magnetic easy axis is locked in the grain.

In this paper we formulate the linearized hydrodynamics appropriate for a small amplitude sound wave propagating in the presence of a strong external magnetic field. The orientation director $\boldsymbol{n}$ is coupled into the equations for density and velocity. The sound velocity is found to be anisotropic, depending on the angle between the propagation vector $\boldsymbol{q}$ and the field $\boldsymbol{H}$. The anisotropy Δ is estimated to be of order 10^{-5} and is frequency dependent. It shows anamolous behaviour at the characteristic frequency ω_c of the orientation and also exhibits relaxation behaviour for large damping.

2. Hydrodynamics

We begin by writing the magnetization density as

$$\boldsymbol{m}(r, t) = (\rho_m(r, t)/\rho_{0m})\, m_0 \boldsymbol{n}(r, t) \tag{1}$$

where ρ_m is the local density of magnetic particles and has an average value ρ_{0m}, and m_0 is the average magnetization throughout the fluid; and for a large external field H, it is independent of H (saturation). It depends on the nature and concentration of magnetic grains. Finally $\boldsymbol{n}$ is a unit vector parallel to $\boldsymbol{m}$. Now $\rho_m = C\rho$ and $\rho_{0m} = C_0\rho_0$ where C is the concentration of magnetic particles and ρ is the total density of the fluid. Then

$$\boldsymbol{m}(r, t) = [m_0 C(r, t)\, \rho(r, t)\, \boldsymbol{n}(r, t)]/C_0\rho_0. \tag{2}$$

We are interested in the case where the fluctuations from equilibrium are small. We write $\rho = \rho_0 + \rho'$, $C = C_0 + C'$ and $\boldsymbol{n} = \boldsymbol{n}_0 + \boldsymbol{n}'$, where ρ', C' and $\boldsymbol{n}'$ are small and $\boldsymbol{n}_0$ is parallel to the external field $\boldsymbol{H}$. Linearizing equation (2) in the primed variables we obtained

$$\boldsymbol{m}(r, t) = m_0\boldsymbol{n}_0 + m_0\, [(\rho'/\rho_0)\, \boldsymbol{n}_0 + (C'/C_0)\, \boldsymbol{n}_0 + \boldsymbol{n}'] = \boldsymbol{m}_0 + \boldsymbol{m}'. \tag{3}$$

The first term $\boldsymbol{m}_0 = m_0\boldsymbol{n}_0$ is the average, constant magnetization in the fluid. The next three terms in equation (3) give the various contributions to the fluctuating part $\boldsymbol{m}'$. The first of them gives the contribution arising from density fluctuations with no change in the concentration. This term is completely determined by ρ' and hence does not lead to new terms in the hydrodynamics. The second gives the contribution arising from concentration fluctuations at constant density. They are described by a diffusion-like equation similar to that in a non-magnetic binary fluid (Mountain and Deutch 1969). Although concentration fluctuations can lead to new terms in the damping of sound waves, their effect on the sound velocity will be presumed to be small. The last term gives the contribution arising from orientation fluctuations of the grains. These fluctuations will be described by an equation for $\boldsymbol{n}$ similar to that used in the description of nematic liquid crystals (Leslie 1968, Stephen 1970, Forster *et al* 1971).

Having determined which variables will enter our hydrodynamical description we must next specify the free energy of the fluid. There are two types of interactions we must consider. The first is the interaction between pairs of magnetic grains. The interaction energy will be of order $U_g \sim \mu M_p$, where μ is the dipole moment of a grain and M_p is the magnetization of the grain. Typically $M_p \sim 500$–1000 (CGS). In the presence of an external magnetic field H there will be an interaction between the grains and the field: $U_H \sim \mu H$. Thus for large fields (10^4 G) $U_H \gg U_g$ and we can ignore the grain–grain interactions. In this case a simple form for the free energy should be valid

$$E = -m_0(\boldsymbol{n} . \boldsymbol{H}). \tag{4}$$

Note that $\boldsymbol{H}$ refers to the external field. In general there are internal fields created by the fluid, but these are of order m_0 and therefore small compared to H for a large enough H.

The linearized hydrodynamical equations are the continuity equation

$$\frac{\partial \rho'}{\partial t}+\rho_0(\nabla . \boldsymbol{v})=0, \tag{5}$$

the conservation of momentum

$$\rho_0 \frac{\partial v_i}{\partial t}+T_{ij,j}=0 \tag{6}$$

and the director equation

$$I \frac{\partial^2 n_i}{\partial t^2}+g_i+\gamma_1 \dot{n}_i{}^*+\gamma_2 n_j d_{ji}=0 \tag{7}$$

where Cartesian tensor notation has been used where necessary. In equations (6) and (7)

$$T_{ij}=p\delta_{ij}+t_{ij} \tag{8}$$

where p is the pressure and t_{ij} is the (Leslie 1968) 'viscosity' tensor. The important terms are

$$t_{ij}=-\alpha_2 n_i \dot{n}_j{}^*-\alpha_3 n_j \dot{n}_i{}^*. \tag{9}$$

Other terms involve the viscosities α_1, α_4, α_5 and α_6 and the velocity gradients. These will not affect the sound velocity to first order, whereas the terms retained in equation (9) will. They would have to be included if we wanted to calculate the damping of sound waves however. We use the notation of Stephen (1970) in equation (9); α_2 and α_3 are reversed in the notation of Leslie (1968). The terms in $\dot{n}_i{}^*$ and d_{ij} are

$$\dot{n}_i{}^*=\frac{\partial n_i}{\partial t}-\tfrac{1}{2}\,[(\nabla x v)\,x\boldsymbol{n}]_i \tag{10}$$

and

$$d_{ij}=\tfrac{1}{2}(v_{i,j}+v_{j,i}). \tag{11}$$

In equation (7), I is the moment of inertia density for the grains and is estimated by $I\sim \rho_0 C_0 a^2$ where a is the characteristic length of the largest magnetic unit able to rotate rigidly under an applied torque. In nematic liquid crystals a is the molecular length, and the inertial term is negligibly small except at very high frequencies (Groupe d'Etude 1969). However, in magnetic fluids the term might be important if long needles form and are not broken up by the sound wave. The director force g_i is given by

$$g_i=\partial E/\partial n_i \tag{12}$$

and equation (7) is to be solved under the constraint $\boldsymbol{n}^2=1$.

Let the external field $\boldsymbol{H}$ be in the z direction. There is an obvious symmetry in the xy plane so that we can take the sound wave as propagating in the xz plane with no loss of generality. Then $v_y=0$ and all gradients in the y direction vanish. Let ϕ be the angle between $\boldsymbol{H}$ and the propagation vector $\boldsymbol{q}$, and let θ be the angle between $\boldsymbol{n}$ and $\boldsymbol{H}$. If we write $n_z=\cos\theta\simeq 1$ and $n_x=\sin\theta\simeq\theta$, then equation (7) reduces to a single equation for the orientation angle θ:

$$I\frac{\partial^2\theta}{\partial t^2}+\frac{\partial\theta}{\partial t}+\tfrac{1}{2}(\lambda+1)\frac{\partial v_z}{\partial x}+\tfrac{1}{2}(\lambda-1)\frac{\partial v_x}{\partial z}+(m_0 H/\gamma_1)\theta=0 \tag{13}$$

where $\lambda=\gamma_2/\gamma_1$.

We next evaluate t_{ij} from equation (9). From conservation of angular momentum and the Onsager reciprocal relations (Parodi 1970) it can be shown that $\gamma_1 = C_0^{-1}(\alpha_2 - \alpha_3)$ and $\gamma_2 = C_0^{-1}(\alpha_2 + \alpha_3)$. Using equation (7) to solve for $\dot{n}_i^*$ in equation (9), and inserting into equation (6) we find that the velocity equations are

$$\rho_0 \frac{\partial v_x}{\partial t} + \frac{\partial p}{\partial x} + \tfrac{1}{2}C_0(\lambda - 1)\frac{\partial J}{\partial z} = 0 \tag{14}$$

$$\rho_0 \frac{\partial v_z}{\partial t} + \frac{\partial p}{\partial z} + \tfrac{1}{2}C_0(\lambda + 1)\frac{\partial J}{\partial x} = 0. \tag{15}$$

Terms involving the velocity gradients have been omitted for the same reason that velocity gradient terms are omitted from equation (9). J is given by

$$J = m_0 H\theta + \frac{I\partial^2\theta}{\partial t^2}. \tag{16}$$

The pressure can be written $p = p_0 + p'$, where p_0 is constant and p' is a small fluctuation. For sound waves we have

$$p' = (\partial p/\partial\rho)_{sc\theta}\ \rho' \quad \text{and} \quad p' \equiv v_0^2\ \rho'. \tag{17}$$

The thermodynamic derivative is evaluated at constant entropy (s), as well as constant orientation and concentration. v_0 is defined by equation (17) and represents the sound velocity in the zero frequency (hydrodynamic) limit. Below we will find corrections to this for finite frequencies which involve the effect of the orientation oscillations.

3. Sound propagation

We look for solutions of the form

$$\rho'(x, z, t) = \sum_q \rho_q \exp\left[i(q_1 x + q_3 z)\right] \exp(\mathrm{i}\omega t) \tag{18}$$

with similar expansions for v_{qx}, v_{qz} and θ. Insertion of equation (18) into equations (5), (13), (14) and (15) leads to four coupled linear equations in ρ_q, v_{qx}, v_{qz} and θ_q. For a nontrivial solution the determinant of the coefficients must vanish and this leads to the dispersion relation

$$\begin{aligned}(1 - (\omega/\omega_c)^2 + \mathrm{i}\omega\tau)(v_0^2 q^2 - \omega^2) \\ = (\mathrm{i}\gamma_1 C_0/4\rho_0\omega)\, q^2 (1 - \omega^2/\omega_c^2)\left[v_0^2 q^2(\lambda\cos 2\phi - 1)^2 - \omega^2(\lambda^2 - 2\lambda\cos 2\phi + 1)\right]\end{aligned} \tag{19}$$

where $\tau = \gamma_1/m_0 H$ is the relaxation time, and $\omega_c = (m_0 H/I)^{1/2}$ is the natural frequency for the orientation fluctuations.

$$q_1 = q\sin\phi \quad \text{and} \quad q_3 = q\cos\phi. \tag{20}$$

For a given ω, equation (19) determines q. For propagating waves we will have $(\gamma_1\omega/\rho_0 v_0^2) \ll 1$ so that $v_0^2 q^2 \simeq \omega^2$. This suggests that we try a perturbation solution to equation (19) of the form

$$q = (\omega/v_0) + q' \tag{21}$$

where q' is small. We linearize equation (19) in q', treating the right-hand side as small.

The solution for q' is

$$-2\omega v_0 q' = \tfrac{1}{4} C_0 \lambda^2 (i\gamma_1\omega/\rho_0 v_0^2)\, \omega^2 (1-\omega^2/\omega_c^2) \times \{[(1-\omega^2/\omega_c^2) - i\omega\tau]/[(1-\omega^2/\omega_c^2)^2 + \omega^2\tau^2]\} \sin^2 2\phi. \tag{22}$$

Note that q' in equation (22) is complex. Writing $q' = q_r' + iq_i'$ and defining the phase velocity as $v = \omega/q_r$ we find that

$$v = v_0(1+\Delta) \tag{23}$$

where Δ is given by

$$\Delta = C_0(\lambda^2/8)\, (\gamma_1\omega/\rho_0 v_0^2)\, (1-\omega^2/\omega_c^2)\, \{\omega\tau/[(1-\omega^2/\omega_c^2)^2 + \omega^2\tau^2]\} \sin^2 2\phi. \tag{24}$$

The speed of sound is thus anisotropic and depends upon the angle ϕ between $\boldsymbol{q}$ and the field $\boldsymbol{H}$. The anisotropy vanishes for $\phi = 0$, $\pi/2$ indicating that the sound wave exerts no torque on the magnetic dipoles and hence there is no interaction with H in these cases.

The anisotropy Δ is also frequency dependent. For low frequencies $\omega\tau \ll 1$, $\omega \ll \omega_c$ and $\Delta \rightarrow 0$. There is no magnetic interaction in this case because the frequency is so slow that θ has time to relax to zero within an oscillation of the wave. This is the equilibrium hydrodynamic limit. There is anamolous behaviour in Δ when $\omega = \omega_c$ and it changes sign at this point. For very large frequencies $\omega\tau \gg 1$, $\omega \gg \omega_c$ and Δ approaches the limiting value Δ_∞. For $\phi = \pi/4$,

$$\Delta_\infty = -C_0(\lambda^2/8)\, (\gamma_1\tau/\rho_0 v_0^2)\, \omega_c^2 = -C_0(\lambda^2/8)\, (\gamma_1^2/\rho_0 v_0^2 I). \tag{25}$$

We can determine the maximum and minimum values of Δ by looking for the roots of $\partial\Delta/\partial\omega = 0$. This yields the quadratic equation

$$(1-\alpha^2)\, x^2 - 2x + 1 = 0 \tag{26}$$

where $x = \omega^2/\omega_c^2$ and $\alpha = \omega_c\tau$. The solution is

$$x = (1 \pm \alpha)/(1-\alpha^2) \qquad (\alpha \neq 1) \tag{27}$$

$$x = \tfrac{1}{2} \qquad (\alpha = 1). \tag{28}$$

Suppose $\alpha \ll 1$, then $x \simeq 1 \pm \alpha$. The root $x = 1-\alpha$ gives a positive value of Δ

$$\Delta_{\max} \propto \alpha(1-\alpha). \tag{29}$$

The root $x = 1+\alpha$ gives a negative value of Δ

$$\Delta_{\min} \propto -\alpha(1+\alpha). \tag{30}$$

The behaviour of $\Delta(x)$ in this case will look like that sketched in figure 1. As α increases the separation in x between $\Delta_{\max}$ and $\Delta_{\min}$ increases. For $\alpha \simeq 1$ there is only one non-zero ($x = \frac{1}{2}$) and this gives a positive value for Δ. Thus, the minimum disappears and goes over to an inflection point so that $\Delta(x)$ looks like that sketched in figure 2. Finally for $\alpha \gg 1$ the only positive root is $x = 1/\alpha$. This gives a maximum independent of α as shown in figure 3.

The case $I = 0$ is appropriate if the needles tend to break up under the influence of the sound wave: then $\omega_c \rightarrow \infty$ and equation (24) reduces to a purely relaxation type of behaviour

$$\Delta(\omega) = (C_0\lambda^2/8)\, (\gamma_1\omega/\rho_0 v_0^2)\, [\omega\tau/(1+\omega^2\tau^2)] \sin^2 2\phi. \tag{31}$$

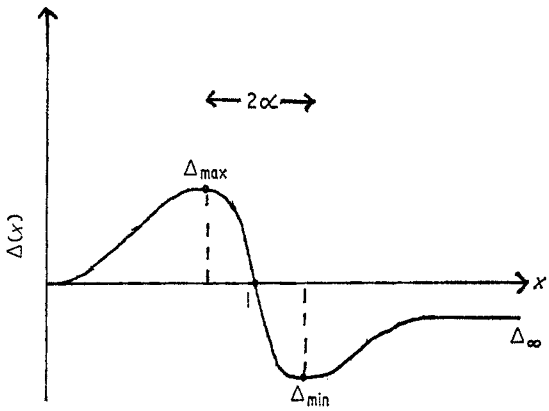

Figure 1 Behaviour of $\Delta(x)$ for $\alpha \ll 1$.

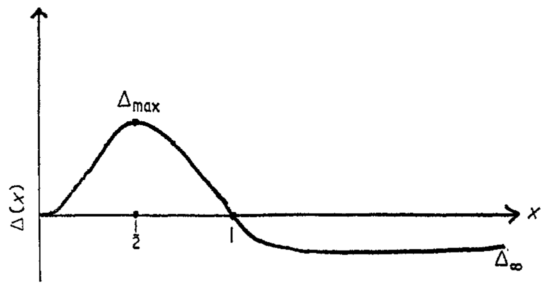

Figure 2. Behaviour of $\Delta(x)$ for $\alpha \simeq 1$.

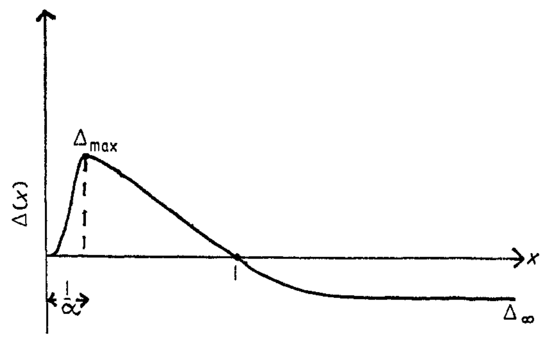

Figure 3. Behaviour of $\Delta(x)$ for $\alpha \gg 1$.

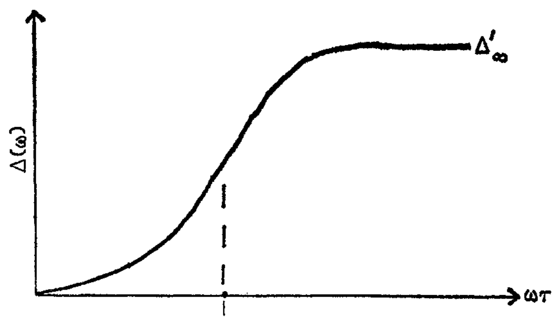

Figure 4. $\Delta(\omega)$ when $I=0$, $\omega_c \rightarrow \infty$.

The frequency dependence is shown in figure 4. Δ is always positive and approaches a constant limit Δ_∞' as $\omega\tau \gg 1$, that is

$$\Delta_\infty' = C_0\lambda^2 (m_0H/8\rho_0v_0^2) \sin^2 2\phi. \tag{32}$$

We can estimate the magnitude of Δ_∞' by putting $C_0 \simeq 1$, $\lambda \simeq 1$, $m_0 \simeq 10^2$–10^3, $H \simeq 10^4$, $\rho_0 \simeq 1$ and $v_0 \simeq 10^5$ all in CGS units. This gives $\Delta_\infty \simeq 10^{-4}$–$10^{-5}$ which is very small but within the resolution of phase-sensitive detection techniques (Moran and Lüthi 1969).

There are several possible contributions to Δ which we have ignored. Inclusion of viscous dissipation would lead to a term in equation (24) of second order in viscosity (Mountain 1966). For low viscosity this should be negligible, but could easily be corrected for if present. Concentration and temperature fluctuations will lead to terms involving additional relaxation. These terms can be separated out from the effect of orientation fluctuations if the relaxation times for the three processes are well separated. Also the coupling for the latter two processes should be much smaller than that due to orientation fluctuations when the external field is large.

The imaginary part of q' in equation (22) leads to the damping of the sound waves, as can be seen from equation (18). It is usual to consider the ratio q_i'/ω^2

$$q_i'/\omega^2 = \tfrac{1}{8}C_0\lambda (\gamma_1/\rho_0v_0^3) \{(1-\omega^2/\omega_c^2)^2/[(1-\omega^2/\omega_c^2)^2 + \omega^2\tau^2]\} \sin^2 2\phi. \tag{33}$$

This represents the damping due to orientation fluctuations and it is also anisotropic. For low frequencies ($\omega\tau \ll 1$, $\omega \ll \omega_c$) and for high frequencies ($\omega\tau \gg 1$, $\omega \gg \omega_c$) it approaches the same limit,

$$\left(\frac{q_i}{\omega^2}\right)_{\max} = \frac{C_0\lambda\gamma_1}{8\rho_0v_0^3} \sin^2 2\phi, \tag{34}$$

whereas it vanishes at $\omega = \omega_c$. Therefore we get symmetric behaviour about $\omega = \omega_c$. If $I \simeq 0$ then $\omega_c \to \infty$ and equation (33) reduces to a form typical of a relaxation process. It has the maximum value, given by equation (34), for $\omega\tau \ll 1$ and decreases monotonically to zero for $\omega\tau \gg 1$. For $\phi = 45°$ the maximum value is about $10^{-16}\gamma_1$, using the same estimates as before. This must be compared to the damping produced by ordinary viscosity, also anisotropic (Forster *et al* 1971), which we have ignored:

$$q_v/\omega^2 \sim \eta/\rho_0v_0^3 \sim 10^{-15}\eta \tag{35}$$

where η is an average viscosity. If $\gamma_1 \sim \eta$, as expected, the damping due to viscosity is about an order of magnitude larger than that due to relaxation of the orientation. Note, however, that q_v/ω^2 is independent of ω, hence it may be possible to observe equation (34) as the dispersive term in the total damping. To the first order, contributions to the damping from concentration and temperature fluctuations will also be nondispersive. Higher-order terms will involve the concentration and temperature relaxation times and will be independent of the external field H.

4. Conclusions

A simple linearized hydrodynamical theory for magnetic fluids in the presence of a strong external magnetic field has been presented. The theory involves two additional variables not present in an ordinary fluid. One is the concentration of magnetic particles and the other is a unit vector (the director) parallel to the local magnetization, which is described by an equation similar to that used to describe orientation fluctuations in nematic liquid crystals. The equations are solved for a sound wave propagating at an angle ϕ from the external field direction. The sound velocity is shown to be anisotropic, depending on the angle ϕ. The anisotropy Δ, which is estimated to be about 10^{-5} for a field of 10^4 G, shows an interesting frequency dependence. The variation with frequency gives information on the size of the needles or chains which remain stable against the perturbing sound wave. It would be interesting to consider the phase velocity and damping of sound near the 'liquid' and 'solid' phase transitions of the grains, since one might expect anomalies in some of the transport coefficients, particularly viscosity. For dilute samples, Brillouin light scattering might be possible. If the needles remain stable additional Brillouin lines due to propagating orientation fluctuations would arise.

References

de Gennes P G and Pincus P A 1973 *Phys. Kodens. Mater.* **34** 197
Forster D, Lubensky T C, Martin P C, Swift J and Pershan P S 1971 *Phys. Rev. Lett.* **26** 1016
Groupe d'Etude des Cristaux Liquides (Orsay) 1969 *J. Chem. Phys.* **51** 816
Hayes C F 1975 *J. Coll. Int. Sci.* **46** at press
Jenkins J T 1972 *Arch. Ratl. Mech. Anal.* 42
Jordan P C 1973 *Molec. Phys.* **25** 261
Leslie F M 1968 *Arch. Ratl. Mech. Anal.* **28** 265
Moran T J and Lüthi B 1969 *Phys. Rev.* **187** 710
Mountain R D 1966 *Rev. Mod. Phys.* **38** 205
Mountain R D and Deutch J M 1969 *J. Chem. Phys.* **50** 1103
Parodi O 1970 *J. Phys.* **31** 581
Rosenweig R E 1966 *Int. Sci. Technol.* **55** 48
Stephen M J 1970 *Phys. Rev.* A2 1558