

Shear-Excited Sound in Magnetic Fluid

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Perceptible sound is shown to be excited in ferrofluids by the shear motion of a rigid plate, if the fluid is exposed to a magnetic field oblique both to the plate and to the direction of propagation. This is in contrast to other fluids, including anisotropic ones such as nematic liquids.

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Ferrofluids, or magnetic fluids, are colloidal suspensions of nanosized ferromagnetic particles stably dispersed in a carrier liquid. Exposed to an external magnetic field, they behave paramagnetically, with susceptibilities χ unusually large for liquids. In combination with slow magnetic responses and general liquid behavior, this causes ferrofluids to display much unexpected behavior, severely testing, and with time perfecting, our understanding of the interplay between hydrodynamics and electromagnetism [1–5]. Equally important, it also opens up a wide range of interesting applications.

In this Letter, we draw attention to the fact that oscillatory shear motion of a wall in ferrofluids will excite sound, which may be picked up by a microphone positioned at the opposite end of the liquid container, say, a few cm away. This is striking, because given the viscosity of the fluid, its field-free response to the wall's shear motions should die out after a few viscous penetration depths, within fractions of a mm, and no signals whatever are expected at the microphone's position.

The amplitude of the predicted, shear-excited sound is proportional to $\Omega\tau M^2 \sin 2\varphi$, where $\Omega \equiv \frac{1}{2}\nabla \times \mathbf{v}$ is the vorticity, and \mathbf{v} the velocity of the fluid; M is the magnetization, τ its relaxation time, and φ the angle between the wall's normal and M 's projection onto the shear plane. When an external field breaks the isotropy of a fluid, coupling between longitudinal and transverse velocity modes is to be expected on general ground. But this formula shows that it is the combination in ferrofluids of the unusually large magnetic susceptibility with its macroscopically slow relaxation ($\chi \sim 1$, $\tau \sim 10^{-4}$ s) that makes the effect under focus measurable and relevant.

Although the emphasis in magnetic fluid research has been on incompressible flow configurations, a fair-sized number of papers [6–13] consider the propagation of sound, mainly focusing on the sound velocity and attenuation, in varying geometries and field configurations. All neglected the field-mediated coupling between sound and shear wave, and assumed pure density waves free of vorticity. This is a generally justified simplification only in ordinary liquids, but not in ferrofluids, in which magnetodissipation is strong.

Magnetodissipation in ferrofluids is said to occur when the actual magnetization $\mathbf{M}(\mathbf{r}, t)$ deviates from its

equilibrium value $\mathbf{M}^{\text{eq}}[\mathbf{H}, \rho, T]$, determined by the local magnetic field $\mathbf{H}(\mathbf{r}, t)$, the local density $\rho(\mathbf{r}, t)$, and the local temperature $T(\mathbf{r}, t)$. Magnetodissipation may be present not only in oscillatory flows (of frequency ω), but also in stationary ones. Starting from the low-frequency end, its effect scales with $\omega\tau$ in the first case and with $\Omega\tau$ in the second case, where τ is the relaxation time of the magnetization. The two best studied magnetodissipative phenomena, both in incompressible flow configurations, are (i) the enhanced shear viscosity ($\sim \Omega\tau$) in a static magnetic field [2,14] and (ii) the reduction of this enhancement in response to a high-frequency ac field (“negative viscosity”) [15–17]. In both cases, the vorticity Ω leads to an off-equilibrium magnetization, $(\mathbf{M} - \mathbf{M}^{\text{eq}}) \neq 0$, which enters the momentum balance, feeding back to the dynamics of Ω and changing the apparent viscosity.

In compressible flow situations such as sound, circumstances are only slightly more complicated. Here deviations of \mathbf{M} from equilibrium are induced not only by the transverse component Ω of the velocity, but also by the longitudinal one, $\nabla \cdot \mathbf{v}$, or, equivalently, by density fluctuations $\delta\rho$. Since any $(\mathbf{M} - \mathbf{M}^{\text{eq}})$, again entering the momentum balance, feeds back to the dynamics of $\nabla \cdot \mathbf{v}$ and Ω simultaneously, these two flow fields become coupled: A finite Ω gives rise to $(\mathbf{M} - \mathbf{M}^{\text{eq}})$, which leads to $\nabla \cdot \mathbf{v} \sim \delta\rho \neq 0$. Vice versa, a finite $\delta\rho \sim \nabla \cdot \mathbf{v}$ leads, again via $(\mathbf{M} - \mathbf{M}^{\text{eq}})$, to $\Omega \neq 0$. Therefore, a sound wave propagating through magnetized ferrofluids is accompanied by vorticity; and an oscillating shear generator excites sound. One may refer to this effect as magnetodissipative coupling between shear and sound, which works, as will become clear soon, only if the orientation of the applied magnetic field is neither parallel nor perpendicular to the direction of propagation (cf. Fig. 1).

To streamline the arguments and work out the physics clearly, we shall implement three realistic simplifications below: (i) weak magnetic field, (ii) hydrodynamic regime $\omega\tau \ll 1$, and (iii) adiabaticity of sound propagation and shear diffusion. It is not difficult to abandon any of these simplifications, but doing so will not qualitatively change the predictions, only obscure them, and unnecessarily and considerably complicate the end formulas. This would be sensible only in direct comparison to experiments, with a specific ferrofluid and a given geometry.

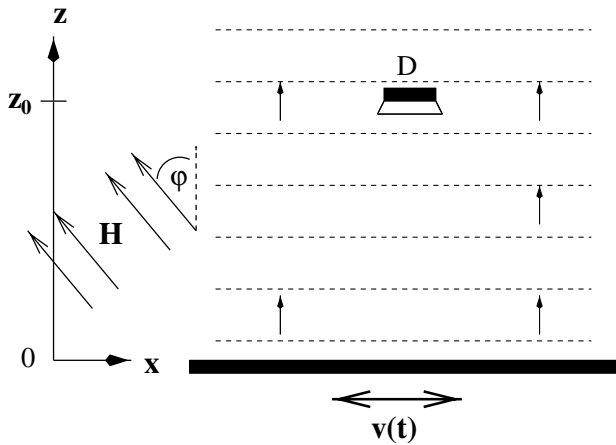


FIG. 1. Given a magnetic field \mathbf{H} in the x - z plane, at an angle φ , a plate oscillating along x is predicted to emit sound waves $\sim \sin 2\varphi$. This may be picked up by a microphone \mathbf{D} located at $z = z_0$, where z_0 is much larger than both the wavelength of sound and the viscous penetration depth. (Filled arrows indicate the direction of propagation, and dashed lines the crests of the sound wave.)

Weak fields, the first simplification, imply the linear constitutive relation, $\mathbf{M}^{\text{eq}} = \chi \mathbf{H}$ and a diagonal form for the Onsager matrix. For a typical ferrofluid, the linear constitutive relation is accurate to within 10% at a field strength of around $H = 5$ kA/m, at which shear-excited sound should be well observable. The 10% inaccuracy is hardly important at present since we are focused on predicting a qualitatively new effect. The same is true of the off-diagonal terms in the Onsager matrix, such as $\lambda_1 M_i (\nabla_j v_j)$ and $\lambda_2 M_j v_{ij}^0$ in the equation of motion for the magnetization \mathbf{M} (see Ref. [5]). These gain importance in relation to the field-independent, diagonal terms for higher fields, but they do not alter the fact that sound and shear couple at finite fields, low or high.

The hydrodynamic regime $\omega\tau \ll 1$ was selected because the considered effect is strong enough here, and the associated calculation remains simple. Adiabaticity or adiabatic limit means that the entropy per unit mass remains constant during the oscillation, $\delta\sigma = 0$, which is to be contrasted with the isothermal limit, $\delta T = 0$. Adiabaticity is valid because in ferrofluids, shear diffusion and sound are usually fast processes on the time scale of heat conduction: The Prandtl number, Pr , given by the quotient of characteristic thermal diffusion time over viscous diffusion time, or, equivalently, by kinematic viscosity over heat diffusivity, $\text{Pr} = \nu/\kappa$, is usually of the order of 10–100. (Depending on the ferrofluid, we have $\nu \approx 10^{-6}$ – 10^{-3} m²/s, and $\kappa \approx 10^{-7}$ – 10^{-5} m²/s.) Adiabaticity means the magnetic susceptibility χ must be taken as a function of σ and ρ , rather than T and ρ .

Now a few words on the equations we start from to deduce the effect of shear-excited sound, since there is still considerable controversy about the valid ferrofluid dynamics. The point is, although for compressible flows, especially sound damping, there is considerable difference

between our version of the ferrofluid dynamics [5] and the other two, by Shliomis [2] and Felderhof [4], all three lead to very similar results in the present context, for the excitation of sound by shear. This is mainly because of two points. First, a large portion of the differences (especially those terms mentioned above, preceded by $\lambda_1, \lambda_2, \dots$) vanish in the weak-field limit; second, the remaining differences are in the field dependence of the eigenvalues of the matrix in Eq. (15), and not in its eigenvectors. The first yields the information on how a magnetic field alters the velocity and damping of sound, a smallish effect; the second yields the information on how sound and shear couple. Note also that the hydrodynamic Maxwell theory [18] would have yielded exactly the same results, since it is completely equivalent to the ferrofluid dynamics of Ref. [5] in the hydrodynamic limit $\tau\omega \ll 1$ (see [19]). This ends the introduction.

We start from the recently derived ferrofluid dynamics [5], a generalized hydrodynamic-type theory that contains the magnetization as a slow variable. The density $\rho(\mathbf{r}, t)$ and the velocity field $\mathbf{v}(\mathbf{r}, t)$ obey their respective conservation laws

$$\partial_t \rho + \nabla_j (\rho v_j) = 0, \quad (1)$$

$$\partial_t (\rho v_i) + \nabla_j (\Pi_{ij} - \Pi_{ij}^{\text{D}}) = 0, \quad (2)$$

with the stress tensors given as

$$\Pi_{ij} = A \delta_{ij} + \rho v_i v_j - H_i B_j - \frac{\mu_0}{2} (h_i M_j - h_j M_i), \quad (3)$$

$$\Pi_{ij}^{\text{D}} = 2\eta_1 v_{ij}^0 + \eta_2 \delta_{ij} (\nabla \cdot \mathbf{v}), \quad (4)$$

where $v_{ij}^0 \equiv \frac{1}{2} (\nabla_i v_j + \nabla_j v_i) - \frac{1}{3} \delta_{ij} (\nabla \cdot \mathbf{v})$, while $A \equiv -u + sT + \mu\rho + \rho v^2 + \mathbf{H} \cdot \mathbf{B}$ is a function of the energy density u , its variables s, ρ, \mathbf{B} , and its conjugate variables T, μ (chemical potential), \mathbf{H} . In the weak-field limit, the relaxation of the magnetization is governed by

$$\partial_t \mathbf{M} + (\mathbf{v} \cdot \nabla) \mathbf{M} - (\mathbf{\Omega} \times \mathbf{M}) = -\chi \mathbf{h} / \tau, \quad (5)$$

where \mathbf{h} , defined generally as $\mu_0^{-1} \partial u / \partial \mathbf{M}$, reduces to $(\mathbf{M} - \mathbf{M}^{\text{eq}}) / \chi$ for linear constitutive relations, with $\mathbf{M}^{\text{eq}} = \chi \mathbf{H}$. (The field variables \mathbf{H}, \mathbf{M} , and \mathbf{B} are given in SI units, with μ_0 the vacuum permeability.) For the evolution of the magnetic field, it is sufficient to take the electric field as static, and employ the Maxwell equations as

$$\nabla \cdot \mathbf{B} = 0, \quad \nabla \times \mathbf{H} = 0. \quad (6)$$

Now, we consider plane wave perturbations of the uniformly magnetized rest state: $\delta\rho, \mathbf{v}, \mathbf{\Omega}, \delta\mathbf{M}, \mathbf{h}, \dots$, all proportional to $e^{i(\mathbf{k} \cdot \mathbf{r} + \omega t)}$, with \mathbf{k} and ω denoting wave vector and frequency. Note the distinction between $\delta\mathbf{M}$ and $\mathbf{h} = (\delta\mathbf{M} - \delta\mathbf{M}^{\text{eq}}) / \chi$, where δ denotes the deviation from the respective homogeneous, motionless rest state. $\delta\mathbf{M}^{\text{eq}} \times (\mathbf{H}, \rho, \sigma)$ is nonvanishing because \mathbf{H} and ρ varies with time. (As discussed above, σ is constant.) While $\delta\mathbf{M}$ is nonvanishing even if the magnetization is perpetually in

equilibrium, or $\delta\mathbf{M} = \delta\mathbf{M}^{\text{eq}}$, we have $\mathbf{h} \neq 0$ only off-equilibrium, when there is magnetodissipation.

An immediate consequence of $\mathbf{M} = \mathbf{B}/\mu_0 - \mathbf{H}$ and Eqs. (6) are the two relations

$$\delta\mathbf{H} = -\delta\mathbf{M}_{\parallel}, \quad \delta\mathbf{B} = \mu_0\delta\mathbf{M}_{\perp}, \quad (7)$$

where \parallel and \perp denote the parallel and perpendicular components with respect to \mathbf{k} , the wave vector. With $\delta\mathbf{M}^{\text{eq}} = \chi\delta\mathbf{H} + \mathbf{H}\delta\chi$, this allows one to express \mathbf{h} as

$$\mathbf{h} = [(1 + \chi)\delta\mathbf{M}_{\parallel} + \delta\mathbf{M}_{\perp} - \mathbf{M}(\chi_{\rho}/\chi)\delta\rho]/\chi, \quad (8)$$

where $\chi_{\rho} \equiv \partial\chi(\rho, \sigma)/\partial\rho$. Equations for the longitudinal and transverse velocity are obtained by taking the divergence and curl of Eq. (2), respectively, giving

$$\left\{ \left[c_0^2 + \frac{\mu_0 M^2}{\rho\chi} \left(\frac{\rho^2 \chi_{\rho}^2}{\chi^2} - \frac{\rho^2 \chi_{\rho\rho}}{2\chi} \right) \right] \nabla^2 - \partial_t^2 \right\} \delta\rho \quad (9)$$

$$+ \frac{4}{3} \frac{\eta_1 + \eta_2}{\rho} \nabla^2 \partial_t \delta\rho - \frac{\mu_0}{\chi} \left(\frac{\rho\chi_{\rho}}{\chi} \right) \mathbf{M} \cdot \nabla^2 \delta\mathbf{M} = 0, \quad (10)$$

$$\rho \partial_t \boldsymbol{\Omega} - (\mu_0/4) \nabla \times \nabla \times (\mathbf{h} \times \mathbf{M}) - \eta_1 \nabla^2 \boldsymbol{\Omega} = 0,$$

where c_0 is the zero-field, adiabatic sound velocity, and

$\chi_{\rho\rho} \equiv \partial^2\chi(\rho, \sigma)/\partial\rho^2$. Let us examine Eqs. (9) and (10), step by step: (i) In the absence of fields ($\mathbf{H}, \mathbf{M} = 0$), the familiar case of a normal liquid is retrieved, with two decoupled modes, the damped propagating sound wave, and the diffusive shear mode. (ii) At finite fields but negligible magnetodissipation ($\mathbf{H}, \mathbf{M} = 0, \mathbf{h} \rightarrow 0$), sound and shear remain decoupled, with sound displaying a shift in its dispersion relation, and shear still unaffected: Inserting Eq. (8) with $\mathbf{h} = 0$ into (9) yields an anisotropic sound velocity $c^2 = c_0^2 + \Delta c^2$, with

$$\Delta c^2 = \frac{\mu_0}{\rho\chi} \left[\left(\frac{\rho\chi_{\rho}}{\chi} \right)^2 \frac{\chi}{1 + \chi} M_{\parallel}^2 - \left(\frac{\rho^2 \chi_{\rho\rho}}{2\chi} \right) M^2 \right]. \quad (11)$$

Using the specifications of a typical ferrofluid, with $\chi \approx 1$, $\rho \approx 1 \text{ g/cm}^3$, $\chi \propto \rho$, and taking $H = 5 \text{ kA/m}$, the relative increment $\Delta c/c_0$ is rather small, less than 10^{-4} .

(iii) Now consider the interesting case of finite magnetodissipation, $\mathbf{h} \neq 0$, which needs to be solved by eliminating $\delta\mathbf{M}$ and \mathbf{h} from Eqs. (9) and (10), in favor of $\delta\rho$ and $\boldsymbol{\Omega}$, by employing Eq. (5). This is most easily accomplished in the low-frequency limit, in which terms of order $(\omega\tau)^2$ and $(\omega\tau)(\Omega\tau)$ may be neglected. Then $\delta\mathbf{M}$ is given as

$$\delta\mathbf{M}_{\parallel} = \left(\frac{\rho\chi_{\rho}}{(1 + \chi)\chi} \right) \frac{\mathbf{M}_{\parallel}}{\rho} \left[1 - \frac{\tau\partial_t}{1 + \chi} \right] \delta\rho + \tau(\boldsymbol{\Omega} \times \mathbf{M})_{\parallel}/(1 + \chi), \quad (12)$$

$$\delta\mathbf{M}_{\perp} = (\chi_{\rho}/\chi) \mathbf{M}_{\perp} [1 - \tau\partial_t] \delta\rho + \tau(\boldsymbol{\Omega} \times \mathbf{M})_{\perp}, \quad (13)$$

with \mathbf{h} given by these expressions and Eq. (8). Inserting all three into Eqs. (9) and (10) yields the set of equations we need. Consider first the coupling term $\nabla \times \nabla \times (\mathbf{h} \times \mathbf{M})$ of Eq. (10), which takes the form

$$\frac{\tau\chi\chi_{\rho}}{1 + \chi} (\mathbf{H}_{\perp} \times \mathbf{H}_{\parallel}) \nabla^2 \partial_t \delta\rho + \nabla \times \nabla \times [(\boldsymbol{\Omega} \times \mathbf{H}) \times \mathbf{H}]. \quad (14)$$

The first term is the interesting one, as it clearly states the fact that given density fluctuations $\delta\rho(t)$, the component of the vorticity $\boldsymbol{\Omega}$ that is parallel to $(\mathbf{H}_{\perp} \times \mathbf{H}_{\parallel})$ —or to $(\mathbf{k} \times \mathbf{H})$ —is also excited. Denoting this component as Ω , the coupled portion of Eqs. (9) and (10) appears as

$$\begin{pmatrix} k^2 c_0^2 - \omega^2 + i\omega k^2 \left(\frac{4}{3} \eta_1 + \eta_2 \right) / \rho + \mathcal{O}(H^2), & i\omega k^2 \mu_0 \tau \chi H_{\parallel} H_{\perp} \chi_{\rho} / (1 + \chi) \\ i\omega k^2 \mu_0 \tau \chi H_{\parallel} H_{\perp} \chi_{\rho} / (1 + \chi), & 4i\omega (i\omega + \eta_1 k^2 / \rho) + \mathcal{O}(H^2) \end{pmatrix} \begin{pmatrix} \nabla \cdot \mathbf{v} \\ \Omega \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}. \quad (15)$$

For $H = 0$, only the diagonal elements remain, and the solvability condition reflects the existence of two independent modes, sound and shear, with the solutions given as (1, 0) and (0, 1). Starting from this as the zeroth order solution, we can calculate the corrections for finite fields. The task is simple if we confine the calculation to order H^2 . At that level of accuracy the field-dependent diagonal terms, abbreviated by $\mathcal{O}(H^2)$, contribute only to the solvability condition, or dispersion relations, while the off-diagonal terms enter only the solution vector; i.e., they alone account for the coupling of the two modes. The latter is an effect of large relative size as it corrects the two zeros in the solution vector. The field-dependent, diagonal terms are longish and their contributions insignificant, that is, the reason they have been left unspecified. Qualitatively, their

contribution is also of order H^2 , where the change in sound velocity, as has been shown below Eq. (11), is negligible. The field-induced sound attenuation scales with the factor $\mu_0 \tau \chi H^2 / \eta_1$; it is more significant but still small. For a ferrofluid with $\tau \approx 0.5 \text{ ms}$, $\eta_1 \approx 0.5 \text{ Pa}$, and $\chi \approx 1$ [20] this factor is, for $H = 5 \text{ kA/m}$, less than 3%. A detailed and systematic investigation of magnetodissipative sound attenuation in ferrofluids will be considered in a separate paper.

We now calculate the corrections to the solution vectors. Starting with the soundlike mode we insert the dispersion

$$k_{\text{sound}} = \pm \left[\frac{\omega}{c_0} - \frac{i}{2} \frac{\omega^2}{\rho c_0^3} \left(\frac{4}{3} \eta_1 + \eta_2 \right) \right] + \mathcal{O}(H^2) \quad (16)$$

into Eq. (15) and obtain the solution

$$\begin{pmatrix} \nabla \cdot \mathbf{v} \\ \Omega \end{pmatrix} = \begin{pmatrix} 1 \\ \alpha \end{pmatrix}, \quad \alpha = \frac{i\omega\tau\mu_0}{4c_0^2} \frac{\chi\rho\chi}{1+\chi} H_{\parallel}H_{\perp}, \quad (17)$$

where we have used the inequality $\omega\eta_i(\rho c_0^2) \ll 1$, valid for acoustic sound. Equation (17) states that sound is accompanied by a finite shear in magnetized ferrofluids if the direction of propagation is neither parallel nor perpendicular to the applied field.

Likewise, inserting the spectrum of the shear mode

$$k_{\text{shear}} = \pm \frac{1-i}{\sqrt{2}} \sqrt{\frac{\omega\rho}{\eta_1}} + \mathcal{O}(H^2), \quad (18)$$

into Eq. (15), we obtain the eigenvector

$$\begin{pmatrix} \nabla \cdot \mathbf{v} \\ \Omega \end{pmatrix}_{\text{shear}} = \begin{pmatrix} -4\alpha \\ 1 \end{pmatrix}. \quad (19)$$

Equation (19) is the key result of this paper, and quite sufficient for predicting the main outcome of the experiment suggested in the introduction. Taking $\Omega = \Omega_0$ and $\nabla \cdot \mathbf{v} = 0$ as the appropriate boundary conditions for the experiment, we can calculate the amplitudes of the two modes by solving $A_{\text{sound}}(1, \alpha) + A_{\text{shear}}(-4\alpha, 1) = (0, \Omega_0)$, yielding

$$A_{\text{shear}} = \Omega_0 + \mathcal{O}(H^4), \quad A_{\text{sound}} = \alpha\Omega_0 + \mathcal{O}(H^4). \quad (20)$$

This result can be tested, as mentioned, by the setup depicted in Fig. 1. A rigid plate, at $z = 0$ and in contact with a magnetized ferrofluid, executes harmonic oscillations in the x - y plane, along y . If the magnetic field $\mathbf{H} = (H_{\perp}, 0, H_{\parallel}) = H(\sin\varphi, 0, \cos\varphi)$ is such that both H_{\perp} and H_{\parallel} are nonzero, a soundlike wave is excited. Because of its weak attenuation, it is detectable by a microphone at a distance far beyond the penetration depth of the shear mode. The associated amplitude of the pressure δp is given by

$$|\delta p| = c_0^2 \delta\rho = c_0^2 \rho \frac{\nabla \cdot \mathbf{v}}{-i\omega} = \frac{\tau\mu_0}{2} \frac{\rho\chi\rho\chi}{1+\chi} H^2 \Omega_0 \sin 2\varphi. \quad (21)$$

For concreteness consider the oscillation frequency $\omega = 2\pi \times 1000$ Hz, and a ferrofluid with the viscosity of $\eta_1 = 0.5$ Pa. Then the penetration depth $\sqrt{\eta_1/(\omega\rho)} \approx 0.3$ mm of the shear mode is less than a mm, while the sound mode displays a spatial decay length of the order of 200 km. Assuming that the shear is generated by a piezoshear crystal, with a deflection amplitude of $\Delta x = 1$ μm , we have the amplitude $\Omega_0 = \omega^{3/2}(\rho/\eta_1)^{1/2}\Delta x = 22.3$ Hz. Applying a magnetic field of $H = 5$ kA/m at an angle of $\varphi = \pi/4$, the pressure amplitude resulting from Eq. (21) is $\delta p \approx 2$ μbar , exceeding the acoustic threshold of the

human ear by more than 3 orders of magnitude. In other words, one should actually be able to hear the shear-generated sound.

Finally, a few remarks on the magnetic susceptibility as used in this paper. Knowing its dependence on density and temperature, $\chi(\rho, T)$, the above employed derivative $\rho\chi_{\rho}(\rho, \sigma)$ may be expressed as

$$\rho \frac{\partial\chi(\rho, \sigma)}{\partial\rho} = \rho \frac{\partial\chi(\rho, T)}{\partial\rho} + T \frac{\partial\chi(\rho, T)}{\partial T} \frac{c_i^2\alpha_v}{C_v}. \quad (22)$$

According to Eq. (22), both magnetostrictive and magnetocaloric contributions are involved, where $\alpha_v = -(1/\rho)\partial\rho(p, T)/\partial T$ denotes the thermal expansion coefficient, c_i the isothermal sound velocity, and $C_v = T\partial\sigma(T, \rho)/\partial T$ the specific heat at constant volume, all evaluated at zero field. Usually, the temperature dependence of the susceptibility is weak at room temperature, and for a typical olefine-based carrier liquid the dimensionless factor $c_i^2\alpha_v/C_v$ can be estimated as 0.3.

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