Anisotropic acoustic waves in rarefied nematic liquid crystals

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Why rarefied nematic liquid crystals?

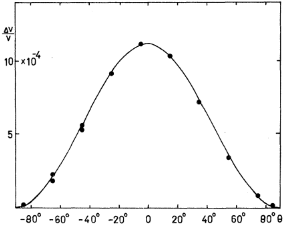


FIG. 2. Angular dependence of sound velocity. $T=21°C$, $\nu=10$ MHz, and $H=5$ kOe. θ is the angle between the field direction and propagation direction. Solid line is $12.5\times10^{-4}\cos^2\theta$.

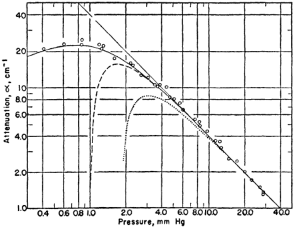


FIG. 1. Attenuation of sound at 1 Mc/sec. in helium. Circles—experimental results. Heavy full line—exact hydrodynamic. Light full line—first approximation, hydrodynamic and Burnett. Dashed line—second approximation, hydrodynamic. Dotted line—second approximation, Burnett.

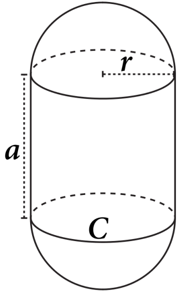
Acoustic waves travel in NLC faster in the direction parallel to the nematic director [MLS72].

First order theory better fits experimental data on acoustic attenuation at low pressure [Gre49].

Curtiss collision operator

In his seminal paper, Curtiss [Cur56] proposed a kinetic theory for spherocylindrical molecules as an idealisation of a polyatomic gas.

- He considered a larger configuration space made by **position**, **velocity**, the **Euler angles** for describing the orientation of each molecule, and the **angular velocity** with respect to a fixed coordinate system.
- Molecules would interact by **excluded volume**, which give rise to **short range interactions**, hence the **nematic ordering**.



This led Curtiss to formulate the following **Boltzmann** type equation,

$$\partial_t f + \nabla_{\boldsymbol{r}} \cdot (\boldsymbol{v} f) + \nabla_{\boldsymbol{\alpha}} \cdot (\dot{\boldsymbol{\alpha}} f) = C[f, f] \qquad (1)$$

where $f(\boldsymbol{r}, \boldsymbol{v}, \boldsymbol{\alpha}, \boldsymbol{\omega})$ is the usual first reduced distribution function and $C[f, f]$ is the collision operator defined as

$$C[f, f] = -\iiiint (f_1' f' - f_1 f)(\boldsymbol{k} \cdot \boldsymbol{g}) S(\boldsymbol{k}) d\boldsymbol{k} d\boldsymbol{v}_1 d\boldsymbol{\alpha}_1 d\boldsymbol{\omega}_1$$

with $S(\boldsymbol{k})d\boldsymbol{k}$ being the surface element of the excluded volume and $\boldsymbol{g} = \boldsymbol{v} - \boldsymbol{v}_1$. Here without loss of generality the equation is stated in **absence of external force** and **torque**.

Collision invariants

It is possible to prove that the following quantities are **collision invariants** for $C[f, f]$, i.e.

$$\iiint \psi^{(i)} C[f, f] d\boldsymbol{v}_1 d\boldsymbol{\omega}_1 d\boldsymbol{\alpha}_1 = 0.$$

- $\psi^{(1)} = 1$, the **number of particles** in the system;
- $\psi^{(2)} = m\boldsymbol{v}$, the **linear momentum**;
- $\psi^{(3)} = \mathbb{I}^1 \cdot \omega + \boldsymbol{r} \times m\boldsymbol{v}$, the **angular momentum**;
- $\psi^{(4)} = \frac{1}{2} m\boldsymbol{v} \cdot \boldsymbol{v} + \frac{1}{2}\boldsymbol{\omega} \cdot \mathbb{I} \cdot \boldsymbol{\omega}$, the **kinetic energy of the system**.

[1] The inertia tensor for the spherocylinder we are considering.

The hydrodynamic equations – notation

We first introduce the **number density**, i.e.

$$n(\boldsymbol{r}) = \iiint f(\boldsymbol{r}, \boldsymbol{v}, \alpha, \boldsymbol{\omega}) d\boldsymbol{v} d\alpha d\boldsymbol{\omega}.$$

Then we can give a meaning to the following *chevrons*, i.e.

$$\langle\langle \cdot \rangle\rangle(\boldsymbol{r}) := \frac{1}{n(\boldsymbol{r})} \iiint \cdot \, f(\boldsymbol{r}, \boldsymbol{v}, \alpha, \boldsymbol{\omega}) d\boldsymbol{v} d\alpha d\boldsymbol{\omega}.$$

Using this notation we can define **macroscopic stream velocity** and **macroscopic stream angular velocity** respectively as:

$$\boldsymbol{v}_0 := \langle\langle \boldsymbol{v} \rangle\rangle, \qquad \boldsymbol{\omega}_0 := \langle\langle \boldsymbol{\omega} \rangle\rangle.$$

The Hydrodynamic Equations – Curtiss Balance Laws

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Testing (1) against the first two **collision invariants** and integrating, Curtiss obtained the following **balance laws**:

$$\partial_t \rho + \nabla_{\boldsymbol{r}} \cdot (\rho \boldsymbol{v}_0) = 0,$$

$$\rho\Big[\partial_t \boldsymbol{v}_0 + (\nabla_{\boldsymbol{r}} \boldsymbol{v}_0)\boldsymbol{v}_0\Big] + \nabla_{\boldsymbol{r}} \cdot (\rho \mathbb{P}) = 0,$$

where ρ is the **density** defined as $\rho(\boldsymbol{r}) = mn(\boldsymbol{r})$ and $\mathbb{P}$ is the **pressure tensor** defined as $\mathbb{P} := \langle\langle \boldsymbol{V} \otimes \boldsymbol{V} \rangle\rangle$, with V being the **peculiar velocity** $\boldsymbol{V} := \boldsymbol{v} - \boldsymbol{v}_0$.

The hydrodynamic equations – surprise balance laws

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For the third collision invariant we took a different route than Curtiss, which led to the following balance law

$$\rho\Big[\partial_t \boldsymbol{\eta} + (\nabla_{\boldsymbol{r}} \boldsymbol{\eta})\boldsymbol{v}_0\Big] + \nabla_{\boldsymbol{r}} \cdot (\rho \mathbb{N}) = \boldsymbol{\xi}, \tag{2}$$

where $\boldsymbol{\eta}$ is the **macroscopic intrinsic angular momentum** defined as $\boldsymbol{\eta}(\boldsymbol{r}) := \langle\!\langle \mathbb{I} \cdot \omega \rangle\!\rangle$ and $\mathbb{N}$ is the **couple tensor** defined as $\mathbb{N} := \langle\!\langle \boldsymbol{V} \otimes (\mathbb{I}\boldsymbol{\omega}) \rangle\!\rangle$. Here ξ_l is defined in tensor notation as $\langle\!\langle mn(\varepsilon_{lki} v_i v_k)\boldsymbol{e}_l \rangle\!\rangle$ and we proved that $\boldsymbol{\xi}$ vanishes (as stated by Curtiss in [Cur56]) in this particular setting.

Maxwell–Boltzmann distribution

In [Cur56] Curtiss gives an expression for the Maxwell–Boltzmann distribution, i.e. the distribution $f^{(0)}$ such that $C[f^{(0)}, f^{(0)}]$ vanishes:

$$f^{(0)}(\boldsymbol{v}, \boldsymbol{\omega}) = n \frac{\sin(\alpha_2) Q}{\int Q \sin(\alpha_2) d\boldsymbol{\alpha}} {\frac{m^{\frac{3}{2}}}{2\pi\theta}}^{3} (\Gamma_1 \Gamma_2 \Gamma_3)^{\frac{1}{2}} \exp\left[-m \frac{|\boldsymbol{V}|}{2\theta} - \frac{\boldsymbol{\Omega} \cdot \mathbb{I} \cdot \boldsymbol{\Omega}}{2\theta} \right],$$

where the **peculiar angular velocity** defined as $\boldsymbol{\Omega} := \boldsymbol{\omega} - \boldsymbol{\omega}_0$, Γ_i are the moments of inertia of the spherocylinder we are considering and $Q := \exp\left[\frac{\boldsymbol{\omega}_0 \cdot \mathbb{I} \cdot \boldsymbol{\omega}_0}{2\theta}\right]$.

Notice in particular that we assumed $\boldsymbol{\omega}_0$ and the **kinetic temperature** $\theta = \frac{m}{2} \boldsymbol{V} \cdot \boldsymbol{V} + \frac{1}{2} \boldsymbol{\Omega} \cdot \mathbb{I} \cdot \boldsymbol{\Omega}$ are fixed.

Momentum closure around the equilibrium

Now we can use the Maxwell–Boltzmann distribution to compute an approximation of the **pressure tensor** near the equilibrium, i.e.

$$\mathbb{P}^{(0)} = \theta \boldsymbol{Id}.$$

We can define the **pressure** as $p = \rho\theta$ and rewrite,

$$\left[\partial_t \boldsymbol{v}_0 + (\nabla_{\boldsymbol{r}} \boldsymbol{v}_0)\boldsymbol{v}_0\right] = -\frac{1}{\rho}\nabla p,$$

which is the well known **Euler equation** that if linearised yields the **wave equation**. **Unfortunately** the same procedure results in a **vanishing** $\mathbb{N}^{(0)}$.

Balance laws for kinetic temperature

We need another way to formulate the **constitutive relation** for the **couple tensor**. We begin by observing that from $\psi^{(4)}$ we get the following balance law:

$$\dot{\psi} + \nabla_{\boldsymbol{r}} \boldsymbol{v}_0 : \mathbb{P} + \nabla_{\boldsymbol{r}} \boldsymbol{\omega}_0 : \mathbb{N} - \nabla \cdot \left[\mathbb{P}^T \boldsymbol{v}_0 + \mathbb{N}^T \boldsymbol{\omega}_0\right] \geq 0$$

where $\psi = \langle\langle \theta \rangle\rangle$. We add $\boldsymbol{\xi}$ and observe that if we integrate with appropriate boundary condition the expression is the **rate of work** theorem that was the starting point of Leslie–Ericksen theory:

$$\dot{\psi} + \nabla_{\boldsymbol{r}} \boldsymbol{v}_0 : \mathbb{P} + \nabla_{\boldsymbol{r}} \boldsymbol{\omega}_0 : \mathbb{N} - \nabla \cdot \left[\mathbb{P}^T \boldsymbol{v}_0 + \mathbb{N}^T \boldsymbol{\omega}_0\right] + \boldsymbol{\xi} \geq 0. \quad (3)$$

Noll–Coleman procedure

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Since we are happy with our **pressure tensor** so far we make the following **ansatz**

$$\psi = \psi(\nu, \nabla\nu)$$

where ν is the **nematic director**. Expanding the total derivative and using the Ericksen identity we get the following expression in tensor notation

$$\dot{\psi} = \varepsilon_{iqp}\Big[\big(\nu_q \frac{\partial\psi}{\partial(\nu_p)} + \partial_k(\nu_q)\frac{\partial\psi}{\partial(\partial_k\nu_p)}\big)\omega_i^0 + \nu_q \frac{\partial\psi}{\partial(\partial_k\nu_p)}\partial_k\omega_i^0\Big] - \frac{\partial\psi}{\partial(\partial_k\nu_p)}\partial_q(\nu_p)\partial(v_q^0)$$

Noll–Coleman procedure

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Substituting this expression into (3) and considering thermodynamic processes for which the exact divergences disappear, we get:

$$\left[\mathbb{P}_{ij} + \frac{\partial \psi}{\partial(\partial_j \nu_p)} \partial_i(\nu_p)\right] \partial_j(\nu_i) + \left[N_{ij} - \varepsilon_{iqp}\nu_q \frac{\partial \psi}{\partial(\partial_j \nu_p)}\right] \partial_j(\omega_i^0)$$
$$\left[P_{pq} - \frac{\partial \psi}{\partial(\partial_p \nu_k) \partial_q(\nu_k)}\right] \varepsilon_{iqp}\omega_i^0 \geq 0.$$

Since the above expression must hold for all thermodynamic processes for which the exact divergences disappear, we get the following **constitutive relations**:

$$\mathbb{P} = \nabla \boldsymbol{\nu}^T \frac{\partial \psi}{\partial(\nabla \boldsymbol{\nu})} + \mathbb{P}^{(0)}, \qquad N_{ij} = \varepsilon_{iqp}\nu_q \frac{\partial \psi}{\partial(\partial_j \nu_p)} = \boldsymbol{\nu} \times \frac{\partial \psi}{\partial(\nabla \boldsymbol{\nu})}.$$

Anisotropic waves

It can be shown that steady spherical solutions of (2) verify $\nabla\boldsymbol{\nu}^T\nabla\boldsymbol{\nu} \approx \boldsymbol{Id} + \boldsymbol{\nu}\otimes\boldsymbol{\nu}$. Therefore for this particular case we have the following choice of **pressure tensor**:

$$\mathbb{P} = \mathbb{P}^{(0)} + \boldsymbol{Id} + \boldsymbol{\nu}\otimes\boldsymbol{\nu}.$$

If we linearise the **Euler equation** with this choice of **pressure tensor** we get the wave equation:

$$\partial_t^2 p - \nabla\cdot\Big[(\langle\!\langle\theta+1\rangle\!\rangle\boldsymbol{Id} + \boldsymbol{\nu}\otimes\boldsymbol{\nu})\nabla p\Big] = 0.$$

Anisotropic waves

It is well known that a planar wave

$$p(\boldsymbol{r}, t) = A\cos(\boldsymbol{k} \cdot \boldsymbol{r} - \omega t)$$

travelling in a transversely isotropic medium has speed of sound

$$\boxed{c_s = (c + cos(\gamma))^2}$$

where γ is the angle between $\boldsymbol{k}$ and $\boldsymbol{\nu}$. A similar reasoning was presented in [BDT14], where a theory for anisotropic waves in **dense liquid** crystals is developed.

Conclusions

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- First order theory arises naturally from Boltzmann type equations, even for rod-like molecules.
- Using a Noll–Coleman argument for the closure of the momentum hierarchy allows us to capture the anisotropy of acoustic waves in rarefied liquid crystals.
- We hope to use the relations that arise from the closure procedure presented today to compute Frank constants from $\mathbb{I}$, the inertia tensor of the spherocylinder we are considering.

Thank you for the attention !

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