PHYSICAL REVIEW A VOLUME 27, NUMBER 2 FEBRUARY 1983

Hydrodynamics of the incommensurate phases of smectics-*E* and $Hg_{3-\delta}AsF_6$

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(Received 16 July 1982)

We discuss the linearized hydrodynamic equations for the incommensurate phases of smectic-*E* liquid crystals and of the mercury chain compound $Hg_{3-\delta}AsF_6$. In particular, we clarify the nature of the spontaneously broken continuous symmetries and point out their consequences for the number of hydrodynamic variables and thus for the number of propagating and diffusive normal modes. In addition, contact is made with a microscopic description by deriving Kubo relations for the transport parameters involved. A discussion of the crossover between propagating and diffusive modes due to finite interactions between the chains and the host lattice of $Hg_{3-\delta}AsF_6$ is also included.

I. INTRODUCTION

During the last few years there has been an increasing interest in incommensurate systems and this development has been reviewed from the theoretical point of view in Ref. 1 and from the experimental point of view (concentrating on resonance experiments) by Blinc.[2] One of the systems which seems to have been investigated most thoroughly for a rather broad variety of its physical properties is the mercury chain compound $Hg_{3-\delta}AsF_6$.[3-6] Another example of an incommensurate system which is supposed to have interesting properties in the domain of low-energy excitations is an incommensurate smectic-*E* liquid crystal which has been identified recently by Brownsey and Leadbetter[7] using x-ray diffraction techniques. A theoretical description of the behavior of an isolated mercury chain (in $Hg_{3-\delta}AsF_6$) has been given by two groups. Emery and Axe[8] used a harmonic chain model leading to a sine-Gordon equation and Mori, Shobu, and Yoshida[9,10] studied the transport behavior of an isolated chain.

In the present paper we will focus our attention exclusively on the three-dimensional hydrodynamic properties of the two systems mentioned above. By hydrodynamic regime we mean that we look at time scales long compared to any microscopic time (e.g., time between two collisions, τ_c), $\omega\tau_c \ll 1$, where ω is the frequency of an excitation, and at length scales which are long compared to any microscopic length scale, l_c of the system, i.e., we have $kl_c \ll 1$, where k is the wave vector of the excitation. To provide a complete hydrodynamic description we will combine the thermodynamic framework (of, e.g., Ref. 11) with the projector formalism of Mori[12] introduced into hydrodynamics by Forster.[13] Using these techniques we can shed new light on the material presented very recently by Axe and one of the authors[14] for $Hg_{3-\delta}AsF_6$. Furthermore we present for the first time a macroscopic description of incommensurate smectic-*E* (Sm-*E*) liquid crystals. In particular, we will clarify why additional normal modes (compared to a crystal) can occur in the systems under consideration.

The paper is organized as follows. In Sec. II we present the hydrodynamics of incommensurate smectic-*E* liquid crystals and propose some experiments to verify the predictions made here. In Sec. III we concentrate on $Hg_{3-\delta}AsF_6$ for temperatures larger than the temperature for the lock-in transition of the mercury chains, and in Sec. IV the behavior of $Hg_{3-\delta}AsF_6$ at temperatures $T < T_c$ is examined. Finally we discuss in the conclusions the relations of our work to the papers on isolated mercury chains and we investigate in some detail the similarities and the differences in the hydrodynamics of the "usual" not incommensurate systems. For incommensurate smectics *E* we will present the details of the hydrodynamic equations whereas for the incommensurate phases of $Hg_{3-\delta}AsF_6$ emphasis is laid on the discussion of the hydrodynamic variables, the spontaneously broken continuous symmetries, and the structure of the normal mode spectrum. The numbers of the coefficients involved in the various tensors describing the dissipative behavior is listed in a table.

II. HYDRODYNAMICS OF INCOMMENSURATE SMECTICS-*E*

In 1980 it was pointed out for the first time by Brownsey and Leadbetter[7] that two incommensurate density waves in one direction can coexist in a smectic-E liquid crystal. In this section we derive the general hydrodynamic equations for such a system and specialize our results for the case of an orthorhombic smectic-E liquid crystal.

To set up the hydrodynamics we have to identify the hydrodynamic variables. In an ordinary smectic-E liquid crystal without incommensurations we have as conserved quantities the total density ρ, the density of linear momentum $\vec{g}$, and the entropy density σ (or equivalently the energy density ϵ). Since smectics-E are believed to be identical to crystal in the hydrodynamic regime, we have as variables characterizing the spontaneously broken translation symmetries three displacement fields $\vec{u}$. Consequently one finds three pairs of propagating modes and two modes which are diffusive (cf. Ref. 11 for the corresponding discussion of the case of a crystal). If we now switch to the incommensurate phase we have two incommensurate density waves in a certain direction (which we will choose to be the z direction, without loss of generality). In the directions transverse to the preferred direction nothing changes when compared to an ordinary smectic-E liquid crystal, i.e., we have as variables the two transverse components of the density of linear momentum and the two transverse displacement fields which are conjugate variables to the two transeverse components of $\vec{g}$, $\vec{g}_t$ ($\vec{g}_t$ serves as generator of the transverse broken translational symmetries). In addition, it is important to notice that the total density ρ is dragged along by $\vec{g}_t$ in these directions. Parallel to the preferred direction, however, the situation is quite different. Due to the incommensurability of the two density modulations along the preferred axis, it becomes possible, by definition of the term incommensurate, to move one density modulation with respect to the other without cost of energy (for infinitesimal displacements). That is there is no finite energy (for $k \to 0$) associated with the relative motion of the two density waves. Correspondingly one has to introduce two densities of linear momentum and two displacement fields parallel to the preferred direction, i.e., it is possible to have more than one spontaneously broken continuous symmetry of the same kind (translational symmetry) in one direction, a feature *unique* to incommensurate systems among all systems studied so far in the hydrodynamic regime.

The possibility to have two densities of linear momentum in certain directions, however, is well known from a rather different field: the case of mixture of immiscible fluids (for a detailed account of the behavior of immiscible fluids in a porous medium, we refer to Ref. 15).

Furthermore, it is necessary to include in the list of conserved quantities the density of one of the two modulations (ρ_A) in addition to the total density ($\rho = \rho_A + \rho_B$, where ρ_B is the density of the other modulation along the preferred direction). Equivalently one might drop the total density as a hydrodynamic variable and keep the two conserved quantities ρ_A and ρ_B separately. Thus we have the following hydrodynamic variables in the incommensurate phase of smectics-E: ρ, the total density, the density ρ_A, the entropy density σ, the two transverse displacement fields $\vec{u}_{\mathrm{tr}}$, the two transverse components of the density of linear momentum $\vec{g}_{\mathrm{tr}}$, two densities of linear momentum parallel to the preferred direction g_i^A and g_i^B, and two displacement fields u_i^A, u_i^B. Correspondingly we expect to find 11 normal modes, diffusive or propagating. To set up the hydrodynamic equations we start as usual (assuming local thermodynamic equilibrium) from the Gibbs relation for the hydrodynamic variables

$$\begin{aligned} d\epsilon = T\,d\sigma - \mu\,d\rho + \phi_{ij}^A d\nabla_i u_j^A + \phi_{ij}^B d\nabla_i u_j^B + \phi_{ij}^{\mathrm{tr}} d\nabla_i u_j^{\mathrm{tr}} \\ + \vec{v}^{\mathrm{tr}} dg^{\mathrm{tr}} + v_i^A dg_i^A + v_i^B dg_i^B + Z^A d\rho^A \;, \end{aligned} \tag{2.1}$$

where T, μ, ϕ_{ij}^A, ϕ_{ij}^B, ϕ_{ij}^{tr}, $\vec{v}^{\mathrm{tr}}$, v_i^A, v_i^B, and Z^A are thermodynamic conjugates as defined in Eq. (2.1). For the pressure we have

$$p = -\epsilon + \mu\rho + Z^A \rho_A + v_i^{\mathrm{tr}} g_i^{\mathrm{tr}} + v_i^A g_i^A + v_i^B g_i^B \;, \tag{2.2}$$

and for the equations of motions for the conserved quantities and the quantities characterizing the broken symmetries,

$$\begin{aligned} &\dot{\rho}_A + \nabla_i j_i^A = 0 \;, \\ &\dot{\rho}_B + \nabla_i j_i^B = 0 \;, \\ &\dot{u}_i^A + X_i^A = 0 \;, \\ &\dot{u}_i^B + X_i^B = 0 \;, \\ &\dot{u}_i^{\mathrm{tr}} + X_i^{\mathrm{tr}} = 0 \;, \\ &\dot{g}_i^{\mathrm{tr}} + \nabla_j \sigma_{ij}^{\mathrm{tr}} = 0 \;, \\ &\dot{g}_i^A + \nabla_j \sigma_{ij}^A = 0 \;, \\ &\dot{g}_i^B + \nabla_j \sigma_{ij}^B = 0 \;, \\ &\dot{\sigma} + \nabla_i j_i^\sigma = \frac{R}{T} \;, \end{aligned} \tag{2.3}$$

where the currents j_i^σ, j_i^A, j_i^B, $\sigma_{ij}^{\mathrm{tr}}$, σ_{ij}^A, σ_{ij}^B and the quasicurrents X_i^A, X_i^B, X_i^{tr} are defined via Eqs. (2.3). As usual R denotes the entropy production. In a first step we relate the thermodynamic conjugate quantities to the hydrodynamic variables. To achieve this it is most convenient to start from the generalized free-energy functional F:

$$
\begin{aligned}
F=\int dV \Bigg[& \frac{(\vec{g}^{\,\mathrm{tr}})^2}{2\rho}+\frac{(g_i^A)^2}{2\rho_A}+\frac{(g_i^B)^2}{2\rho_B}+T\left[\frac{\partial\rho}{\partial T}\right]_p(\delta\rho)(\delta\sigma)+\rho C_p T(\delta\sigma)^2+\chi_1(\delta\rho_A)^2+\chi_2(\delta\rho)(\delta\rho_A) \\
&+\chi_3(\delta\rho_A)(\delta\sigma)+\rho\left[\frac{\partial\rho}{\partial p}\right]_T(\delta\rho)^2+\gamma_{ijkl}(\nabla_i u_j^{\mathrm{tr}})(\nabla_k u_l^{\mathrm{tr}})+\alpha_{ijkl}(\nabla_i u_j^A)(\nabla_k u_l^A) \\
&+\xi_{ijkl}(\nabla_i u_j^A)(\nabla_k u_l^B)+\beta_{ijkl}(\nabla_i u_j^B)(\nabla_k u_l^B)+\zeta_{ijkl}(\nabla_i u_j^{\mathrm{tr}})(\nabla_k u_l^B)+\delta_{ijkl}(\nabla_i u_j^{\mathrm{tr}})(\nabla_k u_l^A) \\
&+\chi_{ij}(\delta\sigma)(\nabla_i u_j^{\mathrm{tr}})+\widetilde{\chi}_{ij}(\delta\rho_A)(\nabla_i u_j^{\mathrm{tr}})+\widetilde{\widetilde{\chi}}_{ij}(\delta\rho)(\nabla_i u_j^{\mathrm{tr}})+\psi_{ij}(\delta\rho_A)(\nabla_i u_j^A)+\widetilde{\psi}_{ij}(\delta\rho)(\nabla_i u_j^A) \\
&+\widetilde{\widetilde{\psi}}_{ij}(\delta\sigma)(\nabla_i u_j^A)+\lambda_{ij}(\delta\rho_A)(\nabla_i u_j^B)+\widetilde{\lambda}_{ij}(\delta\rho)(\nabla_i u_j^B)+\widetilde{\widetilde{\lambda}}(\delta\sigma)(\nabla_i u_j^B)+\tau_1(\partial_x u_z^A-\partial_x u_z^B)^2 \\
&+\tau_2(\partial_x u_z^A+\partial_z u_x)^2+\tau_3(\partial_x u_z^A-\partial_x u_z^B)(\partial_x u_z^A+\partial_z u_x)+\tau_4(\partial_y u_z^A-\partial_y u_z^B)^2 \\
&+\tau_5(\partial_y u_z^A+\partial_z u_y)^2+\tau_6(\partial_y u_z^A-\partial_y u_z^B)(\partial_y u_z^A+\partial_z u_y)\Bigg] \,. \qquad (2.4)
\end{aligned}
$$

The thermodynamic conjugates are then found simply by taking partial derivatives with respect to one quantity while keeping fixed all other variables (representd by the ellipsis in the subscript):

$$
\begin{aligned}
&T=\left.\frac{\delta F}{\delta\sigma}\right|_{\dots}, \quad \mu=\left.\frac{\delta F}{\delta\rho}\right|_{\dots}, \quad Z^A=\left.\frac{\delta F}{\delta\rho^A}\right|_{\dots}, \\
&\vec{v}^{\,\mathrm{tr}}=\left.\frac{\delta F}{\delta\vec{g}^{\,\mathrm{tr}}}\right|_{\dots}, \quad v_i^A=\left.\frac{\delta F}{\delta g_i^A}\right|_{\dots}, \quad v_i^B=\left.\frac{\delta F}{\delta g_i^B}\right|_{\dots}, \\
&\phi_{ij}^B=\left.\frac{\delta F}{\delta\nabla_i u_j^B}\right|_{\dots}, \quad \phi_{ij}^A=\left.\frac{\delta F}{\delta\nabla_i u_j^A}\right|_{\dots}, \\
&\phi_{ij}^{\mathrm{tr}}=\left.\frac{\delta F}{\delta\nabla_i u_j^{\mathrm{tr}}}\right|_{\dots}. \qquad (2.5)
\end{aligned}
$$

To complete the derivation of the hydrodynamic equations we have to relate in a second step the reversible and irreversible parts of the currents introduced via the balance equations (2.3) to the thermodynamic conjugate quantities presented in Eqs. (2.4), and (2.5). In detail we find for the reversible currents ($e_i\equiv$unit vector parallel to the z direction)

$$j_i^{\sigma^R}=\sigma v_j\delta_{ij}^{\mathrm{tr}}+(\sigma_A v_j^A+\sigma_B v_j^B)e_ie_j \,, \qquad (2.6)$$

$$j_i^{A^R}=\rho_A v_j^A e_ie_j+\rho_A v_i^{\mathrm{tr}} \,, \qquad (2.7)$$

$$j_i^{B_R}=\rho v_j^B e_ie_j+\rho_B v_i^{\mathrm{tr}} \,, \qquad (2.8)$$

$$\sigma_{ij}^{\mathrm{tr}^R}=p\delta_{ij}^{\mathrm{tr}}+\delta_{ik}^{\mathrm{tr}}\phi_{kj}^{\mathrm{tr}} \,, \qquad (2.9)$$

$$\sigma_{ij}^{A^R}=p_A e_ie_j+e_ie_k\phi_{kj}^A \,, \qquad (2.10)$$

$$\sigma_{ij}^{B^R}=p_B e_ie_j+e_ie_k\phi_{kj}^B \,, \qquad (2.11)$$

$$X_i^A=g_i^A\rho_A^{-1} \,, \qquad (2.12)$$

$$X_i^B=g_i^B\rho_B^{-1} \,, \qquad (2.13)$$

$$X_i^{\mathrm{tr}}=g_i^{\mathrm{tr}}(\rho_A+\rho_B)^{-1} \,, \qquad (2.14)$$

where

$$g_i^A=\rho_A v_i^A, \;\; g_i^B=\rho_B v_i^B, \;\; g_i^{\mathrm{tr}}=(\rho_A+\rho_B)v_i^{\mathrm{tr}} \,. \qquad (2.15)$$

Some of these equations show quite remarkable features which are unique to incommensurate systems and have not been found for any other hydrodynamic system considered so far. From Eqs. (2.9)–(2.11), e.g., it becomes obvious that the hydrostatic pressure, which must be isotropic, picks up two contributions from the two densities of linear momentum parallel to the preferred direction, where as nothing changes compared to crystals in the directions perpendicular to e_i. Thus we arrive at $p_A+p_B=p$. Even for superfluid systems it has not been necessary to split up the pressure. A quite analogous effect occurs for the reversible current of the entropy density [Eq. (2.5)] where we have to split up the density σ into the contributions coming from the components A and B which are dragged along with the velocities v_A and v_B, respectively. A cross term, which would be allowed by pure symmetry considerations (as behavior under parity or time reversal), has to be excluded in order to guarantee the incommensurability of the system. Or in other words, the structure of the current [Eq. (2.6)] is a direct consequence of the term incommensurability. The same reasoning applies to the quantities g_A and g_B listed in Eq. (2.15) and to the reversible currents of the concentrations ρ_A and ρ_B [Eqs. (2.7) and (2.8)]. The irreversible currents may be derived in a convenient way from the dissipation function R, a quantity which is positive by definition. After having obtained the general structure of R as a function which is quadratic in the thermodynamic conjugates, the irreversible currents are the derivatives of R with respect to the gradients of the thermodynamic conjugate quantities. In detail we have

$$\begin{aligned}R=\int dV[&\kappa_{ij}(\nabla_i T)(\nabla_j T)+\zeta'_{ij}(\nabla_e\phi^{\mathrm{tr}}_{ie})(\nabla_k\phi^{\mathrm{tr}}_{jk})+\zeta''_{ij}(\nabla_e\phi^{B}_{ie})(\nabla_m\phi^{B}_{jm})+\zeta'''_{ij}(\nabla_e\phi^{A}_{ie})(\nabla_m\phi^{A}_{im})\\&+\zeta^{\mathrm{iv}}_{ij}(\nabla_e\phi^{A}_{ie})(\nabla_m\phi^{B}_{jm})+\eta_{ijkl}(\nabla_i v^{\mathrm{tr}}_j)(\nabla_k v^{\mathrm{tr}}_l)+\eta'_{ijkl}(\nabla_i v^{A}_j)(\nabla_k v^{B}_l)+\eta''_{ijkl}(\nabla_i v^{B}_j)(\nabla_k v^{B}_l)\\&+\eta'''_{ijkl}(\nabla_i v^{A}_j)(\nabla_k v^{\mathrm{tr}}_l)+\eta^{\mathrm{iv}}_{ijkl}(\nabla_i v^{A}_j)(\nabla_k v^{A}_l)+\eta^{\mathrm{v}}_{ijkl}(\nabla_i v^{\mathrm{tr}}_j)(\nabla_k v^{B}_l)+\xi'_{ij}(\nabla_l\phi^{\mathrm{tr}}_{ie})(\nabla_j T)\\&+\xi''_{ij}(\nabla_e\phi^{A}_{ie})(\nabla_j T)+\xi'''_{ij}(\nabla_l\phi^{B}_{il})(\nabla_j T)+\psi'_{ij}(\nabla_i Z^A)(\nabla_j Z^A)+\psi''_{ij}(\nabla_i Z^B)(\nabla_j Z^B)\\&+\psi'''_{ij}(\nabla_i Z^A)(\nabla_j Z^B)+\rho'_{ij}(\nabla_i Z^B)(\nabla_j T)+\rho''_{ij}(\nabla_i Z^A)(\nabla_j T)+\nu'_{ik}(\nabla_i Z^B)(\nabla_j\phi^{\mathrm{tr}}_{kj})\\&+\nu''_{ik}(\nabla_i Z^B)(\nabla_j\phi^{B}_{kj})+\nu'''_{ik}(\nabla_i Z^B)(\nabla_j\phi^{A}_{kj})+\nu^{\mathrm{iv}}_{ik}(\nabla_i Z^A)(\nabla_j\phi^{\mathrm{tr}}_{kj})+\nu^{\mathrm{v}}_{ik}(\nabla_i Z^A)(\nabla_j\phi^{A}_{kj})\\&+\nu^{\mathrm{vi}}_{ik}(\nabla_i Z^A)(\nabla_j\phi^{B}_{kj})+\Gamma]\ ,\end{aligned} \tag{2.16}$$

$$\begin{aligned}\Gamma\equiv{}&\tilde\tau(\partial_x v^A_z-\partial_x v^B_z)^2+\tilde\tau_2(\partial_x v^A_z+\partial_z v_x)^2+\tilde\tau_3(\partial_x v^A_z-\partial_x v^B_z)(\partial_x v^A_z+\partial_z v_x)\\&+\tilde\tau_4(\partial_y v^A_z-\partial_y v^B_z)^2+\tilde\tau_5(\partial_y v^A_z+\partial_z v_y)^2+\tilde\tau_6(\partial_y v^A_z-\partial_y v^B_z)(\partial_y v^A_z+\partial_z v_y)\ .\end{aligned}$$

For the irreversible currents we have then

$$\begin{aligned}&\left[j^B_i\right]^I=\frac{\delta R}{\delta\nabla_i Z^B}\bigg|_{\dots}\ ,\quad \left[j^A_i\right]^I=\frac{\delta R}{\delta\nabla_i Z^A}\bigg|_{\dots}\ ,\quad \left[j^\sigma_i\right]^I=\frac{\delta R}{\delta\nabla_i T}\bigg|_{\dots}\ ,\\&\left[X^A_i\right]^I=\frac{\delta R}{\delta\nabla_j\phi^A_{ij}}\bigg|_{\dots}\ ,\quad \left[X^B_i\right]^I=\frac{\delta R}{\delta\nabla_j\phi^B_{ij}}\bigg|_{\dots}\ ,\quad \left[X^{\mathrm{tr}}_i\right]^I=\frac{\delta R}{\delta\nabla_j\phi^{\mathrm{tr}}_{ij}}\bigg|_{\dots}\ ,\\&\left[\sigma^A_{ij}\right]^I=\frac{\delta R}{\delta\nabla_i v^A_j}\bigg|_{\dots}\ ,\quad \left[\sigma^B_{ij}\right]^I=\frac{\delta R}{\delta\nabla_i v^B_j}\bigg|_{\dots}\ ,\quad \left[\sigma^{\mathrm{tr}}_{ij}\right]^I=\frac{\delta R}{\delta\nabla_i v^{\mathrm{tr}}_j}\bigg|_{\dots}\ .\end{aligned} \tag{2.17}$$

We have written down in Eq. (2.16) the terms for g^{tr}_i, g^A_i, g^B_i, etc., separately explicitly in order to show the intricate couplings brought along by the incommensurability. In Table I we have listed the number of coefficients involved in the various viscous tensors for an orthorhombic lattice [which is quite common for smectics-E (Ref. 16)]. In order to exploit the complete set of linearized hydrodynamic equations given above we discuss first the structure of the normal modes (specializing to an orthorhombic structure).

For the reversible part of the spectrum we find to lowest order in k ($\delta\sigma\equiv0$): (a) for $k_y=k_z=0$, $k_x\neq0$,

$$\begin{aligned}&\omega_1^2+\omega_2^2=[\rho_A^{-1}(\tau_1+\tau_2+\tau_3)+\rho_B^{-1}\tau_1]k_x^2\ ,\\&\omega_1^2\omega_2^2=(\rho_A\rho_B)^{-1}k_x^4[\tau_1(\tau_1+\tau_2+\tau_3)-(\tau_1+\tau_3)^2]\ ,\end{aligned} \tag{2.18}$$

TABLE I. Number of parameters that enter the dissipative tensors for orthorhombic incommensurate smectics-E and $Hg_{3-\delta}AsF_6$ above the lock-in transition.

$Hg_{3-\delta}AsF_6$	1 parameter:	β''_{ij}
	2 parameters:	$\kappa_{ij},\beta_{ij},\beta'_{ij},\chi'_{ij},\chi''_{ij},\zeta'_{ij},\xi'_{ij},\psi'_{ij},\psi''_{ij},\tilde\psi_{ij},\tilde\psi'_{ij},\tilde\psi'''_{ij}$
	3 parameters:	γ_{ijkl}
	6 parameters:	η_{ijkl}
Smectics-E	1 parameter:	$\zeta''_{ij},\zeta'''_{ij},\zeta^{\mathrm{iv}}_{ij},\xi''_{ij},\xi'''_{ij},\nu''_{ik},\nu'''_{ik},\nu^{\mathrm{v}}_{ik},\nu^{\mathrm{vi}}_{ik},\eta'_{ijkl},\eta''_{ijkl},\eta^{\mathrm{iv}}_{ijkl}$
	2 parameters:	$\zeta'_{ij},\xi'_{ij},\nu'_{ik},\nu^{\mathrm{iv}}_{ik},\eta'''_{ijkl},\eta^{\mathrm{v}}_{ijkl}$
	3 parameters:	$\kappa_{ij},\psi'_{ij},\psi''_{ij},\psi'''_{ij},\rho'_{ij},\rho''_{ij}$
	4 parameters:	η_{ijkl}

for the modes formed by g_A, g_B, u_A, u_B which are decoupled to lowest order form the other variables. In addition, we get a biquadratic equation in ω for the reversible motion of $u_x, u_y, g_x, g_y, \rho_A, \rho_B$ and two modes $\omega=0$: (b) for $k_z \neq 0, k_x = k_y = 0$ the modes for u_x, g_x and u_y, g_y are decoupled from the rest and we have

$$\omega_1^2 = (\rho_A + \rho_B)^{-1} \tau_2 k_z^2 ,$$
$$\omega_2^2 = (\rho_A + \rho_B)^{-1} \tau_5 k_z^2 . \tag{2.19}$$

For the motion of u_z^A, u_z^B, g_z^A, g_z^B, ρ_A, and ρ_B we get a biquadratic equation in ω coupling these six variables, two modes which are purely diffusive, i.e., $\omega=0$, and two pairs of propagating modes.

Thus we have in either case four pairs of propagating modes. As it is easily checked the same holds for arbitrary directions of the wave vector; the structure, however, becomes more involved because all modes are coupled together and one has to solve a fourth-order equation in ω^2. Since no detailed experiments have been reported so far on the hydrodynamic modes of incommensurate smectics-*E* we refrain from writing this rather opaque expression. Thus we find one additional pair of propagating modes in the incommensurate phase; this fact is intimately connected with the existence of an additional broken symmetry and of an additional density of linear momentum. As expected from general hydrodynamic considerations we find that the dissipation is always proportional to k^2 (both for purely diffusive and for propagating normal modes).

Using the projector technique of Mori applied to hydrodynamics by Forster it becomes possible to connect the equations derived above to expressions containing correlation or absorptive response functions in the hydrodynamic regime. And these quantities can be either determined by experiment (light scattering, neutron scattering, etc.) or evaluated approximately by purely microscopic techniques like, e.g., the equation of motion. Here we concentrate on the presentation of Kubo relations which allow us to establish a connection between the transport parameters involved and the small wave number, small frequency limit of the absorptive response functions. For a detailed account of the formalism we refer to Refs. 13, 17, and 18. After having derived the ingredients of the projector approach in the hydrodynamic regime (matrix of static susceptibilities, frequency matrix, and memory matrix) it is well established (cf., e.g., Ref. 18) how to proceed directly from these expressions to the final Kubo relations without using the explicit results for the absorptive response functions in the hydrodynamic regime (which look unwieldy in the present system, due to the intricate couplings involved).

We find for the Kubo relations which are different in the incommensurate phase (when compared to usual smectics-*E* or crystals)

$$\zeta'' = \lim_{\omega\to 0}\lim_{k\to 0} \omega \chi''_{u_B u_B}(k,\omega) ,$$
$$\zeta''' = \lim_{\omega\to 0}\lim_{k\to 0} \omega \chi''_{u_A u_A}(k,\omega) ,$$
$$\zeta^{\text{iv}} = \lim_{\omega\to 0}\lim_{k\to 0} \omega \chi''_{u_A u_B}(k,\omega) , \tag{2.20}$$

$$\eta^{\text{iv}} = \lim_{\omega\to 0}\lim_{k_z\to 0}\lim_{k_{x,y}\to 0} \frac{\omega}{k^2} \chi''_{g_z^A g_z^A}(k,\omega) ,$$
$$\tilde{\tau}_1 + \tilde{\tau}_2 + \tilde{\tau}_3 = \lim_{\omega\to 0}\lim_{k_x\to 0}\lim_{k_{y,z}\to 0} \frac{\omega}{k^2} \chi''_{g_z^A g_z^A}(k,\omega) ,$$
$$\eta'' = \lim_{\omega\to 0}\lim_{k_z\to 0}\lim_{k_x,k_y\to 0} \frac{\omega}{k^2} \chi''_{g_z^B g_z^B}(k,\omega) ,$$
$$-\tilde{\tau}_3 - \tilde{\tau}_1 = \lim_{\omega\to 0}\lim_{k_x\to 0}\lim_{k_y,k_z\to 0} \frac{\omega}{k^2} \chi''_{g_z^A g_z^B}(k,\omega) ,$$
$$\eta' = \lim_{\omega\to 0}\lim_{k_z\to 0}\lim_{k_x,k_y\to 0} \frac{\omega}{k^2} \chi''_{g_z^A g_z^B}(k,\omega) ,$$
$$\eta_{yxyx} = \lim_{\omega\to 0}\lim_{k_y\to 0}\lim_{k_x,k_z\to 0} \frac{\omega}{k^2} \chi''_{g_x g_x}(k,\omega) ,$$
$$\eta_{xxxx} = \lim_{\omega\to 0}\lim_{k_x\to 0}\lim_{k_y,k_z\to 0} \frac{\omega}{k^2} \chi''_{g_x g_x}(k,\omega) ,$$
$$\xi'_x = \lim_{\omega\to 0}\lim_{k\to 0} \frac{\omega}{k} \chi''_{\sigma u_x}(k,\omega) , \tag{2.21}$$
$$\xi''' = \lim_{\omega\to 0}\lim_{k\to 0} \frac{\omega}{k} \chi''_{\sigma u_B}(k,\omega) ,$$
$$\xi'' = \lim_{\omega\to 0}\lim_{k\to 0} \frac{\omega}{k} \chi''_{\sigma u_A}(k,\omega) ,$$
$$\nu''' = \lim_{\omega\to 0}\lim_{k\to 0} \frac{\omega}{k} \chi''_{\rho_B u_A}(k,\omega) ,$$
$$\nu^{\text{vi}} = \lim_{\omega\to 0}\lim_{k\to 0} \frac{\omega}{k} \chi''_{\rho_A u_B}(k,\omega) .$$

Thus it might become possible to determine experimentally some of the new viscosities involved by using light-scattering techniques, supplementing the information which can be gained studying the dissipation of the sound attenuation in specific directions.

III. HYDRODYNAMICS OF $Hg_{3-\delta}AsF_6$ ABOVE T_c

At temperatures above $T_c = 120$ K the mercury chain compound $Hg_{3-\delta}AsF_6$ is known to form an interesting incommensurate phase[3–6,14] in which the AsF_6 anions form a body-centered tetragonal lattice which is penetrated by two nonintersecting orthogonal arrays of mercury cations parallel to the basal plane edges of the AsF_6 lattices. In Refs. 8–10 these mercury chains have been dealt with as one-dimensional fluids and in Refs. 14 a continuum-type description of the compound as a whole has been presented starting from a Hamiltonian which consists of the elastic energy for four displacement fields and a corresponding expression for the kinetic energy. Here we will focus on the characterization of the hydrodynamic variables and the long-wavelength, low-frequency properties of $Hg_{3-\delta}AsF_6$ for $T > T_c$.

From the hydrodynamic point of view the two arrays of mercury chains give rise to two additonal densities ρ^1_{Hg} and ρ^2_{Hg} (aside from the density of the AsF_6 lattice, ρ). Furthermore, we have two additional densities of linear momentum parallel to the two arrays of mercury chains, reflecting the fact that the mercury chains can be moved with respect to the "host" lattice of AsF_6 anions without cost of energy for $k \to 0$. These two densities of linear momentum g^x_{Hg} and g^y_{Hg} for the two separate arrays of mercury chains (we have taken the z direction to be orthogonal to any mercury chain) have to be supplemented by the three components of the density of linear momentum of the AsF_6 lattice $\vec{g}$ and by three displacement fields $\vec{u}$ for the AsF_6 lattice which characterize the three broken translational symmetries of this lattice. Finally we have to take into account the entropy density σ. Thus we have a total of 12 hydrodynamic variables in $Hg_{3-\delta}AsF_6$ above T_c: three variables characterizing the spontaneously broken continuous translational symmetries $\vec{u}$ and nine conserved quantities $\rho,\rho^1_{\text{Hg}},\rho^2_{\text{Hg}},\sigma,\vec{g},g^x_{\text{Hg}},g^y_{\text{Hg}}$.

Since we have presented the procedure to set up the hydrodynamic equations in detail in Sec. II (for the incommensurate phase of smectics-*E*) we concentrate on some special features; we use for the displacement fields $\vec{u}$ the same notation as in Ref. 14. For the free energy we find

$$F=\int dV \Bigg[\chi_1[(\delta\rho^1_{\text{Hg}})^2+(\delta\rho^2_{\text{Hg}})^2]+\chi_2[(\delta\rho^1_{\text{Hg}})(\delta\rho^2_{\text{Hg}})]+\chi_3[(\delta^1_{\text{Hg}})+(\delta\rho^2_{\text{Hg}})]\delta\rho$$
$$+\chi_4[(\delta\rho^1_{\text{Hg}})+(\delta\rho^2_{\text{Hg}})]\delta\sigma+\chi_5(\delta\sigma)^2+\chi_6(\delta\sigma)(\delta\rho)+\chi_7(\delta\rho)^2+\frac{\vec{g}^2}{2\rho}+\frac{(g^x_{\text{Hg}})^2}{2\rho^1_{\text{Hg}}}+\frac{(g^y_{\text{Hg}})^2}{2\rho^2_{\text{Hg}}}$$
$$+c_{ijkl}(\nabla_i u_j)(\nabla_k u_l)+\lambda_{ij}(\delta\sigma)(\nabla_i u_j)+\tilde{\lambda}_{ij}(\delta\rho)(\nabla_i u_j)+\psi_1[(\delta\rho^1_{\text{Hg}})(\nabla_x u_x)+(\delta\rho^2_{\text{Hg}})(\nabla_y u_y)]$$
$$+\psi_3[(\delta\rho^1_{\text{Hg}})+(\delta\rho^2_{\text{Hg}})](\nabla_z u_z)+\psi_2[(\delta\rho^1_{\text{Hg}})(\nabla_y u_y)+(\delta\rho^2_{\text{Hg}})(\nabla_x u_x)] \Bigg] . \tag{3.1}$$

In Eq. (3.1) c_{ijkl} contains the same terms as presented in Eq. (4.5d) of Ref. 14 and $\psi_1,\ldots,\psi_3$ as well as $\chi_1,\ldots,\chi_7$ have been chosen to satisfy the symmetry properties of a tetragonal lattice symmetry; λ_{ij} contains two independent parameters:

$$\lambda_{ij}=\lambda_1(e_x e_x+e_y e_y)+\lambda_2(\delta_{ij}-e_x e_x-e_y e_y) .$$

As is easily checked, ϵ^B_{xx} and ϵ^B_{yy} of Ref. 14 correspond to ρ^1_{Hg} and ρ^2_{Hg}, whereas quantities like $\epsilon^B_{xy},\epsilon^B_{yz},\epsilon^B_{xz}$ have no analog in our description. In Ref. 14 contributions involving these terms have been deleted using the argument that the Hg chains support no shear strains; in the present description it is obvious from the outset that ρ^1_{Hg} and ρ^2_{Hg} are scalar conserved quantities. All terms involving $\delta\sigma,\delta\rho(\lambda_i,\tilde{\lambda}_{ij}, \chi_3,\ldots,\chi_7)$, however, have been deleted in Ref. 14, whereas general symmetry considerations clearly allow for the existence of these terms (cf. Refs. 11 and 19 for a discussion of corresponding terms in liquid crystals and crystals). For the normal modes the discussion can be carried over (for the most part) from Ref. 14.

For $q_x \neq 0$, $q_y = q_z = 0$ we find the same expression as Eqs. (4.8) and (4.9) of Ref. 14. The two longitudinal modes, however, are formed by six variables: u_x ρ^1_{Hg}, g_x, g^{Hg}_x, and $\delta\sigma,\delta\rho$. These six variables lead to a biquadratic equation in ω^2 and two modes $\omega = 0$, i.e., a total of eight propagating modes and four diffusive modes are found.

For $q_z \neq 0$, $q_x = q_y = 0$ we find expressions identical to Eqs. (4.10) and (4.11) of Ref. 14. In addition, we find one longitudinal propagating mode and two modes which are diffusive. These four modes are formed by u_z, g_z, σ, and ρ. Thus we find the modified Cauchy relation[14] of the transverse mode propagating in the x direction and polarized along the z axis, and the mode propagating in the z direction and polarized in the x direction to hold as well in the present description; i.e., we have shown that this relation must *hold rigorously in the hydrodynamic regime* and is not influenced by the variables ρ and σ which have not been taken into account in the continuum-type description of Ref. 14. For general directions we find five pairs of propagating modes and two diffusive ones (compared with four propagating pairs and four diffusive for $q_x \neq 0$, $q_y = q_z = 0$ and with three propagating pairs and six diffusive for $q_z \neq 0, q_x = q_y = 0$). The crossover between propagating and diffusive modes found in the present system resembles most closely that discussed for biaxial discotics.[19]

The dissipation function for $\mathrm{Hg}_{3-\delta}\mathrm{AsF}_6$ for $T > T_c$ takes the form

$$\begin{aligned} R = \int dV \{ & \kappa_{ij}(\nabla_i T)(\nabla_j T) + \eta_{ijkl}(\nabla_i v_j)(\nabla_k v_l) + \beta_{ij}(\nabla_i v_y^{\mathrm{Hg}})(\nabla_j v_y^{\mathrm{Hg}}) + \beta'_{ij}(\nabla_i v_x^{\mathrm{Hg}})(\nabla_j v_x^{\mathrm{Hg}}) \\ & + \beta''_{ij}(\nabla_i v_x^{\mathrm{Hg}})(\nabla_j v_y^{\mathrm{Hg}}) + \gamma_{ijkl}(\nabla_i v_j)(\nabla_k v_l^{\mathrm{Hg}}) + \chi'_{ij}(\nabla_i T)(\nabla_j Z^1_{\mathrm{Hg}}) + \chi''_{ij}(\nabla_i T)(\nabla_j Z^2_{\mathrm{Hg}}) \\ & + \zeta'_{ij}(\nabla_k \phi_{ik})(\nabla_l \phi_{jl}) + \xi'_{ij}(\nabla_k \phi_{ik})(\nabla_j T) + \psi'_{ij}(\nabla_k \phi_{ik})(\nabla_j Z^1_{\mathrm{Hg}}) + \psi''_{ij}(\nabla_k \phi_{ik})(\nabla_j Z^2_{\mathrm{Hg}}) \\ & + \widetilde{\psi}_{ij}(\nabla_i Z^1_{\mathrm{Hg}})(\nabla_j Z^2_{\mathrm{Hg}}) + \widetilde{\psi}'_{ij}(\nabla_i Z^1_{\mathrm{Hg}})(\nabla_j Z^1_{\mathrm{Hg}}) + \widetilde{\psi}'''_{ij}(\nabla_i Z^2_{\mathrm{Hg}})(\nabla_j Z^2_{\mathrm{Hg}}) + \tau_1(\partial_x v_y^{\mathrm{Hg}} + \partial_y v_x^{\mathrm{Hg}})^2 \\ & \tau_2[(\partial_x v_z + \partial_z v_x^{\mathrm{Hg}})^2 + (\partial_z v_y^{\mathrm{Hg}} + \partial_y v_z)^2] \} , \end{aligned} \tag{3.2}$$

where

$$\beta_{ij} \equiv \beta'_{ij}, \quad \chi'_{ij} \equiv \chi''_{ij}, \quad \psi'_{ij} \equiv \psi''_{ij}, \quad \widetilde{\psi}'_{ij} \equiv \widetilde{\psi}'''_{ij} .$$

The number of the coefficients involved in the various tensors together with corresponding results for the dissipation of smectics-E and $\mathrm{Hg}_{3-\delta}\mathrm{AsF}_6$ below T_c is listed in Table I. As in Sec. II, one can derive Kubo relations for all transport coefficients involved. Here we list only a few of them which will be used in the discussion below:

$$\psi'_\perp = \lim_{\omega \to 0} \lim_{k_\perp \to 0} \lim_{k_\parallel \to 0} \frac{\omega}{k} \chi''_{u_x \rho^1_{\mathrm{Hg}}}(k,\omega) , \tag{3.3}$$

$$\psi'_\parallel = \lim_{\omega \to 0} \lim_{k_\parallel \to 0} \lim_{k_\perp \to 0} \frac{\omega}{k} \chi''_{u_x \rho^1_{\mathrm{Hg}}}(k,\omega) , \tag{3.4}$$

$$\gamma_{xxxx} = \lim_{\omega \to 0} \lim_{k_x \to 0} \lim_{k_y, k_z \to 0} \frac{\omega}{k^2} \chi''_{g_x g_x^{\mathrm{Hg}}}(k,\omega) , \tag{3.5}$$

$$\chi'_\parallel = \lim_{\omega \to 0} \lim_{k_\parallel \to 0} \lim_{k_\perp \to 0} \frac{\omega}{k^2} \chi''_{\sigma \rho^1_{\mathrm{Hg}}}(k,\omega) , \tag{3.6}$$

$$\beta'' + \tau_1 = \lim_{\omega \to 0} \lim_{k_x, k_y \to 0} \lim_{k_\parallel \to 0} \frac{\omega}{k^2} \chi''_{g_y^{\mathrm{Hg}} g_x^{\mathrm{Hg}}}(k,\omega) , \tag{3.7}$$

$$\widetilde{\psi}_\parallel = \lim_{\omega \to 0} \lim_{k_\parallel \to 0} \lim_{k_\perp \to 0} \frac{\omega}{k^2} \chi_{\rho^1_{\mathrm{Hg}} \rho^2_{\mathrm{Hg}}}(k,\omega) , \tag{3.8}$$

where, e.g., $k_x, k_z \to 0$ means $k_x - k_z \to 0$, etc.

Finally we sketch which of the results presented so far have to be modified if a small coupling between the mercury chains and the host lattice is taken into account. One reason for the existence of such a coupling can be the screened Coulomb interaction, but many other mechanisms could be imagined. If this interaction is sufficiently small it can be treated as a perturbation in the hydrodynamic approach [like, e.g., the magnetic dipole interaction in the superfluid phases of ^{3}He (Ref. 18)]. That is, we assume that we have still to deal with the same set of macroscopic variables. Among those, however, only $g_x + g_x^{\mathrm{Hg}}$ and $g_y + g_y^{\mathrm{Hg}}$ are strictly hydrodynamic, whereas $g_x - g_x^{\mathrm{Hg}}$ and $g_y - g_y^{\mathrm{Hg}}$ are, at best, macroscopic variables which relax, due to the interaction, with a finite but long relaxation time τ_c. This approach makes sense if τ_c is very large compared to any macroscopic time scale of the system. For the equations of motion of $g_1 = g_x + g_x^{\mathrm{Hg}}$, $g_2 = g_y + g_y^{\mathrm{Hg}}$, $g_3 = g_x - g_x^{\mathrm{Hg}}$, and $g_4 = g_y - g_y^{\mathrm{Hg}}$ we have

$$g_1 + \nabla_j \sigma_{1j} = 0 , \tag{3.9}$$

$$g_2 + \nabla_j \sigma_{2j} = 0 , \tag{3.10}$$

$$g_3 + \nabla_j \sigma_{3j} = \frac{v}{\tau} , \tag{3.11}$$

$$g_4 + \nabla_j \sigma_{4j} = \frac{v}{\tau} . \tag{3.12}$$

The source term v in Eqs. (3.11) and (3.12) characterizes the strength of the interaction. The structure of all other conservation and quasiconservation laws remains unchanged. Of course, it is no longer necessary to split up the pressure and the entropy current. The consequences of the source term in Eqs. (3.11) and (3.12) on the mode structure, however, is quite drastic. Instead of five pairs of propagating modes we are left with three pairs of propagating modes. In addition, we have now two purely diffusive modes of the structure $\omega \sim i\Gamma k^2$ (Γ is anisotropic and mostly due to the conserved quantities $\delta\rho^1_{\mathrm{Hg}}$ and $\delta\rho^2_{\mathrm{Hg}}$) and two microscopic modes $\omega \sim i\Delta$ (i.e., modes with a finite gap $\Delta \sim v$ at $k = 0$). Thus we predict a crossover of modes for the mercury chain compounds as a function of the frequency: We can have either three (ω very small) or five (ω larger) pairs of propagating modes. The frequency ω_c for which the crossover occurs is directly proportional [cf. Eqs. (3.11) and (3.12)] to the strength of the interaction between host lattice and mercury chains.

IV. HYDRODYNAMICS OF $Hg_{3-\delta}AsF_6$, $T < T_c$

Below a critical temperature T_c a phase transition (which is probably discontinuous[20]) takes place which leads to a locking of the mercury chains, i.e., it takes energy to slide the x system with respect to the v system.[14] Furthermore the symmetry is lowered from tetragonal to orthorhombic (1,1,0) and (1,−1,0) are no longer equivalent. From the hydrodynamic point of view the variables ρ, $\vec{g}$, and $\vec{u}$ characterizing the AsF_6 system remain unchanged. Due to the lock-in transition, however, there is only one density ρ_{Hg} associated with the mercury chains. Furthermore, the Hg chains bring along now only one additional component of density of linear momentum g_{Hg}, because only along the (110) direction the mercury chains can be moved with respect to the AsF_6 lattice without cost of energy for $k \rightarrow 0$. In addition, it is known that the mercury chains show now long-range positional order (contrary to the quasifluid behavior above T_c).

Correspondingly we have to introduce a displacement field w which is parallel to the (1,1,0) direction and which characterizes the spontaneously broken translational symmetry of the mercury chains. Accordingly we have a total of 11 hydrodynamic variables in $Hg_{3-\delta}AsF_6$ below T_c: four variables characterizing broken symmetries $\vec{u},w$ and seven conserved quantities g_{Hg}, ρ_{Hg}, ρ, $\vec{g}$, and σ, the entropy density. For convenience we choose the following coordinate system: $(1,1,0)\equiv x$, $(1,-1,0)\equiv y$, and $(0,0,1)\equiv z$. These coordinates take into account explicitly the preferred direction in the plane. After having made this identification of the coordinate axes it becomes evident that $Hg_{3-\delta}AsF_6$ ($T < T_c$) is isomorphic to the incommensurate phase of smectics-E in the hydrodynamic regime and all conclusions can be carried over immediately.

V. CONCLUSIONS

We have presented the general linearized hydrodynamic equations appropriate for the low-frequency, long-wavelength description of the incommensurate phases of smectic-E liquid crystals and the mercury chain compound $Hg_{3-\delta}AsF_6$. Some of the features presented, however, are clearly not restricted to the system discussed but will occur for all incommensurate systems. Among these we mention the necessity to split up the pressure parallel to the directions showing incommensurations and one further feature unique to incommensurate systems, namely, the structure of the entropy current which consists of various contributions. In addition, it seems worthwhile to notice that incommensurate systems provide us with the first example where two spontaneously broken continuous symmetries of the same kind (broken translational symmetry) can occur along certain directions. The possibility of having additional densities of linear momentum, which reflects the fact that the subsystems can be moved with respect to each other without cost of energy for $k \rightarrow 0$, gives rise to additional normal modes. A rather unique feature, namely, the so-called modified Cauchy relation for the mercury chain compound above the lock-in transition, which has been put forward by Axe and one of the authors, has been established rigorously in the hydrodynamic regime without any restrictions.

Furthermore, we have found for the first time in a hydrodynamic system that the static susceptibilities and the irreversible currents can depend on the antisymmetric parts of the gradients of the displacement and velocity fields, respectively. Throughout the paper we considered the system as three-dimensional and composite. For the mercury chains in $Hg_{3-\delta}AsF_6$ more microscopic techniques have been put forward by Emery and Axe, and by Mori, Shobu, and Yoshida leading to interesting predictions such as singular heat conduction, etc. We believe, however, that these results only hold for truly one-dimensional systems and we expect them to be modified (to a smaller or larger extent, depending on the quantity under examination) if the real problem of a composite, three-dimensional problem must be faced. From our results for the Kubo relation, e.g., it is obvious that there are dissipative couplings between the mercury chains and the AsF_6 lattice and we think it will be very interesting to test experimentally the order of magnitude of these couplings. In addition, symmetry considerations allow for various couplings in the free energy and these couplings are not excluded by the term incommensurable whose consequences show up most clearly in the hydrodynamic regime in the reversible currents. These additional static and irreversible couplings which occur in the general thermodynamic formulation presented here will certainly allow for a better explanation of experiments in the hydrodynamic regime and a deeper understanding of incommensurate systems.

ACKNOWLEDGMENTS

Both of us (H.B. and P.B.) have been supported in part by National Science Foundation Grant No. PHY77-27084. H.B. is also supported by the Deutsche Forschungsgemeinschaft.

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