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Fluctuations in a Fluid Under a Stationary Heat Flux. III. Brillouin Lines

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We present a general theory of the Brillouin lines in a fluid subject to a strong stationary heat flux. The combined effects of sound-absorbing walls and of large spatial inhomogeneities induced by the temperature gradient are computed for the first time. Nonequilibrium sound modes, constructed by WKB techniques, are used. No restrictions have to be made in the theory concerning the scattering geometry and the thermal equations of state.

KEY WORDS: Nonequilibrium stationary state; correlation function; light scattering; Brillouin lines; nonequilibrium sound modes; sound bending; acoustic admittance; mode coupling.

1. INTRODUCTION

In previous work[1,2] we derived from fluctuating hydrodynamics a theory for the correlation functions of the hydrodynamic variables in a fluid far from thermal equilibrium. In particular, we studied a fluid exposed to a stationary heat flux. We derived formal expansions of the correlation functions in terms of the nonequilibrium hydrodynamic modes, i.e., the eigenmodes of the hydrodynamic operator obtained by linearizing the hydrodynamic equations around the nonequilibrium stationary state.

As an application, we computed the central or Rayleigh line of the light-scattering spectrum for (incident) frequencies in both the optical and microwave regimes.[3] The Rayleigh line is generated by the viscoheat modes. These are slow modes evolving on a time scale slow compared to processes associated with sound propagation.

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In this paper, we continue our study of the light-scattering spectrum of nonequilibrium fluids with the Brillouin lines. They are caused by the so-called fast part of the density–density correlation function, i.e., the part that can be represented in terms of the sound modes. In this sense, the present paper is complementary to Ref. 3. However, we do not aim to present a description of the fast part of the complete correlation matrix, nor do we discuss the microwave regime here. Rather, we restrict ourselves to a general theory of the Brillouin lines in the optical regime.

The early theories[4–9] of the Brillouin lines for fluids exposed to a heat flux are only valid near equilibrium (i.e., to linear order in the temperature gradient) and in the limit of large systems. However, it was noticed soon[10,11] that finite-size effects may be important in the systems that have been studied experimentally.[12,13] In particular, Satten and Ronis[10] proposed a linear theory incorporating boundary effects, which considered partially and totally sound absorbing walls in conjunction with fluctuations on the boundaries. They were able to explain the experiments of Beysens *et al.*[12] experiments qualitatively. Kirkpatrick *et al.*[14] were the first to try a nonlinear theory by taking into account the spatial inhomogeneities induced by large temperature gradients. Explicit results for the Brillouin lines were obtained for special thermodynamic equations of state and scattering geometries. Although the nonlinear theory of Kirkpatrick *et al.* does not include boundary effects, their results are also consistent with the measured[12] asymmetry of the Brillouin lines.

In this paper, the boundary effects and the spatial inhomogeneities are taken into account at the same time. Moreover, we do not have to make any assumptions concerning the equations of state or the scattering geometry. We thus present here for the first time a general, nonlinear theory of the Brillouin lines in a nonequilibrium fluid.

In developing the theory, basically two technical problems arose: First, we had to compute explicitly the nonequilibrium sound modes. They are the natural entities to be used in a formal mode expansion of the correlation functions far from equilibrium. The construction of these modes by means of WKB techniques is by itself an elaborate task that will be discussed in a separate publication.[15] Therefore, here we will only state the results. The second problem was the summation of the expansions in terms of these nonequilibrium modes in closed form. This problem also can be treated, within consistent approximations, analytically. This somewhat delicate calculation will be presented in greater detail here. The final expression for the Brillouin lines appears in a form that is physically a very intuitive generalization of the earlier linear theories, which takes into account both the partial or total sound absorption at the walls and the spatial inhomogeneities in the fluid.

The plan of this paper is as follows: In Section 2, we sketch the derivation of a formal expansion of the Brillouin lines in terms of non-equilibrium sound modes, based on fluctuating hydrodynamics. In the non-equilibrium stationary state, one finds mode-coupling contributions by pairs of nonequilibrium sound modes that are responsible for the asymmetry of the Brillouin lines. Then, in Section 3, we summarize the results for the nonequilibrium sound modes, as obtained from our WKB calculation.(15) They involve space-dependent, complex wave vectors, which contain all the relevant information about the profiles of the hydrodynamic fields in the given stationary state. The nonequilibrium sound modes are used in Section 4 to evaluate the oscillator strengths occurring in the mode expansion of the Brillouin lines. In Section 5, finally, we are ready to present our general result for the Brillouin lines in a fluid subject to a stationary heat flux, and we also give a brief interpretation of the result in physical terms. In order not to interrupt the logical line in the derivation of our theory by lengthy calculations, we devote Appendices A–E to technical details.

In a separate paper,(17) we give a more elaborate discussion of our theory, including a comparison with existing theories and experiments, and proposals for nex experiments.

2. MODE EXPANSION OF THE BRILLOUIN LINES

We consider a simple fluid in a gravity field $\mathbf{g} = -g\mathbf{e}_z$ confined between two horizontal (infinite) plates located at $z = -d/2$ and $z = d/2$, which have uniform temperatures T_1 and T_2, respectively. Assuming that the fluid is in a nonconvective stationary state, the macroscopic flow velocity $\mathbf{u}$ vanishes, and the pressure and the temperature have one-dimensional profiles $p(z)$ and $T(z)$, which are the solution of the nonlinear equations

$$\begin{aligned} &\frac{dp}{dz} + g\rho(p, T) = 0 \\ &\frac{d}{dz}\lambda(p, T)\frac{dT}{dz} = 0 \end{aligned} \tag{2.1}$$

with boundary conditions $T(-d/2) = T_1$, $T(d/2) = T_2$. In (2.1), $\rho(p, T)$ and $\lambda(p, T)$ are the mass density and the thermal conductivity, respectively, expressed in terms of p and T via the local thermodynamic equations of state. For the further development of the theory it is not necessary to know the solution $p(z)$, $T(z)$ explicitly. Thus, we assume they are given.

In the actual fluid, there are always fluctuations around the stationary state. They give rise to light scattering.[18] The quantity measured in a light-scattering experiment is the dynamic structure factor, which is basically the Fourier transform of the density–density correlation function.[18] Denoting by $\delta\rho(\mathbf{r}, t)$ the density fluctuation at point $\mathbf{r}$ and time t, and using the symmetry of the problem, the density–density correlation function is given by[1]

$$M_{\rho\rho}(r_{\|}, z_1, z_2; t) = \langle \delta\rho(\mathbf{r}_1, t_1)\, \delta\rho(\mathbf{r}_2, t_2) \rangle_{\text{ss}} \tag{2.2}$$

where $\mathbf{r}_{\|} = (x_1 - x_2, y_1 - y_2)$, $r_{\|} = |\mathbf{r}_{\|}|$, and $t = t_1 - t_2$, and the average is over the stationary state ensemble. Assuming that the scattering volume is centered at point $\mathbf{R}$ (away from the boundaries) and has linear dimensions of the order L_s, one has for the dynamic structure factor[2,16,18]

$$S(\mathbf{k}, \omega; R_z) = \frac{1}{L_s} \iint_{R_z - L_s/2}^{R_z + L_s/2} dz_1\, dz_2\, e^{-ik_z(z_1 - z_2)} \hat{M}_{\rho\rho}(k_{\|}, z_1, z_2; \omega) + \text{c.c.} \tag{2.3}$$

where c.c. denotes the complex conjugate, $\mathbf{k}_{\|} = (k_x, k_y)$, and

$$\hat{M}_{\rho\rho}(k_{\|}, z_1, z_2; \omega) = \int_0^\infty dt \int d\mathbf{r}_{\|} \{\exp[-i(\mathbf{k}_{\|} \cdot \mathbf{r}_{\|} - \omega t)]\}\, M_{\rho\rho}(\mathbf{r}_{\|}, z_1, z_2; t) \tag{2.4}$$

In (2.3), we have further assumed that the scattering volume is uniformly illuminated during a time T_s with $k_x, k_y \gg L_s^{-1}$ and $\omega \gg T_s^{-1}$.

As shown in Ref. 1, one can derive a theory for the density–density correlation function in the stationary state from fluctuating hydrodynamics. To this purpose, one must consider the full correlation matrix

$$\mathsf{M}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = \langle \delta\mathbf{a}(\mathbf{r}_1, t_1)\, \delta\mathbf{a}(\mathbf{r}_2, t_2) \rangle_{\text{ss}} \tag{2.5}$$

where $\delta\mathbf{a} = (\delta p, \delta T, \delta\mathbf{u})$ denotes the fluctuations of all the independent hydrodynamic fields, namely pressure, temperature, and flow velocity, respectively. From (2.5) one obtains in particular $M_{\rho\rho}(r_{\|}, z_1, z_2; t)$, since the fluctuations in density are related to those in pressure and temperature via the thermodynamic identity $\delta\rho = (\gamma/c^2)\, \delta p - \rho\alpha\, \delta T$, where c is the speed of sound, $\gamma = c_p/c_v$ is the ratio of the specific heat (per unit mass) at constant pressure and constant volume, and α is the thermal expansion coefficient.

For times $t_1 > t_2$ the correlation matrix obeys the evolution equation[1,19]

$$\frac{\partial}{\partial t_1} \mathsf{M}(\mathbf{r}_1, t_1; \mathbf{r}, t_2) = -\mathscr{H}(z_1) \cdot \mathsf{M}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) \qquad (t_1 > t_2) \tag{2.6}$$

where the hydrodynamic operator $\mathscr{H}(z)$ is obtained by linearizing the hydrodynamic equations around the stationary state solution $p(z)$, $T(z)$, $\mathbf{u} = 0$. To solve Eq. (2.6), one needs as initial condition the equal-time correlation matrix $\mathsf{M}(\mathbf{r}_1, t_2; \mathbf{r}_2, t_2)$. This is given by[1,19]

$$\mathsf{M}(\mathbf{r}_1, t_2; \mathbf{r}_2, t_2) = \mathsf{A}^{(0)}(z_1)\, \delta(\mathbf{r}_1 - \mathbf{r}_2) + \mathsf{D}(\mathbf{r}_1, \mathbf{r}_2) \tag{2.7}$$

where the first term on the right-hand side is the local equilibrium correlation matrix,[2,16] while $\mathsf{D}(\mathbf{r}_1, \mathbf{r}_2)$ is a long-range nonequilibrium contribution that must be determined from the equation[1,19]

$$\mathscr{H}_{\alpha\gamma}(z_1)\, D_{\gamma\beta}(\mathbf{r}_1, \mathbf{r}_2) + \mathscr{H}_{\beta\gamma}(z_2)\, D_{\alpha\gamma}(\mathbf{r}_1, \mathbf{r}_2) = -C_{\alpha\beta}(\mathbf{r}_1, \mathbf{r}_2) \tag{2.8}$$

Here $\mathsf{C}(\mathbf{r}_1, \mathbf{r}_2)$ is the so-called mode-coupling matrix to be given explicity in Eq. (2.10) below.

In computing $S(\mathbf{k}, \omega; R_z)$ according to Eqs. (2.3)–(2.8), one can make use of three small parameters that occur naturally in the problem as ratios of four characteristic lengths. These lengths are[2,16]

1. The system size d.
2. The macroscopic length L_∇ measuring the scale on which the macroscopic fields appearing in $\mathscr{H}(z)$ vary.
3. The wavelength k^{-1}.
4. The kinetic length L_m, i.e., the ratio of the generalized diffusion coefficients (kinematic viscosity, longitudinal viscosity, or thermal diffusivity) to the speed of sound.

The three small parameters are

$$\varepsilon_1 = L_m k, \qquad \varepsilon_2 = 1/L_\nabla k, \qquad \varepsilon_3 = 1/kd \tag{2.9}$$

They typically all lie between 10^{-4} and 10^{-3}. In the following, we will denote by ε any of the three small parameters.

Since ε is so small, the dynamic structure factor need only be computed to leading order in ε. For this purpose we have to take into account only the zeroth-order part of the mode-coupling matrix, i.e., $\mathsf{C}(\mathbf{r}_1, \mathbf{r}_2) = \mathsf{C}^{(0)}(z_1)\,\delta(\mathbf{r}_1 - \mathbf{r}_2)$ with[1,2,16]

$$\mathsf{C}^{(0)} = k_B\, dT/dz \begin{pmatrix} 0 & 0 & c^2\mathbf{e}_z \\ 0 & 0 & [(\alpha T + \gamma - 1)/\alpha\rho]\,\mathbf{e}_z \\ c^2\mathbf{e}_z & [(\alpha T + \gamma - 1)/\alpha\rho]\,\mathbf{e}_z & 0 \end{pmatrix} \tag{2.10}$$

Although only the leading order of $S(\mathbf{k}, \omega; R_z)$ is asked for, we must treat the operator $\mathscr{H}(z)$, which governs the dynamics of the correlation matrix, up to first order in ε, since the dynamic structure factor contains resonant peaks around the frequencies $\omega \approx 0$ [Rayleigh line $S_H(\mathbf{k}, \omega; R_z)$] and $\omega \approx \pm ck$ [Brillouin lines $S_{B\pm}(\mathbf{k}, \omega; R_z)$]. These frequencies are proportional to the possible energy transfer between the fluid and the electromagnetic field in the scattering process.[18]

In solving Eqs. (2.6) and (2.8) for the correlation matrix, we apply a spectral decomposition of $\mathscr{H}(z)$ in terms of its eigenmodes. Denoting by s the eigenvalues of $\mathscr{H}(z)$, and by $\mathbf{a}^R(\mathbf{r})$ and $\mathbf{a}^L(\mathbf{r})$ the right and left eigenvectors, respectively, the eigenvalue equations are

$$\begin{aligned} \mathscr{H}(z) \cdot \mathbf{a}^R(\mathbf{r}) &= s\mathbf{a}^R(\mathbf{r}) \\ \mathscr{H}^+(z) \cdot \mathbf{a}^L(\mathbf{r}) &= s\mathbf{a}^L(\mathbf{r}) \end{aligned} \tag{2.11}$$

where $\mathscr{H}^+(z)$ is the adjoint operator in the scalar product $(\mathbf{a}_1, \mathbf{a}_2) = \int d\mathbf{r}\, \mathbf{a}_1(\mathbf{r}) \cdot \mathbf{a}_2(\mathbf{r})$. In diagonalizing $\mathscr{H}(z)$, only the eigenvalues must be determined up to first order in ε, because they govern the dynamics of the correlations near the resonances. For the eigenvectors, which determine the amplitudes of the correlations, the zeroth order is sufficient.

Since we restrict ourselves to the Brillouin lines here, we have to consider only the sound modes, i.e., the fast modes for which the eigenvalues are of the order $|s| \approx ck$. These are characterized by three indices $(\sigma, \mathbf{q}_\parallel n)$: $\sigma = +$ or $-$ distinguishes the sound modes with eigenvalues having positive or negative imaginary parts, respectively, $\mathbf{q}_\parallel = (q_x, q_y)$ is a continuous horizontal wave vector, and n is a discrete index associated with a discrete wavenumber q_{nz} for the vertical direction, in which the system is finite. Using a time-scale perturbation theory to separate the slow and fast variables in $\mathscr{H}(z)$, one finds for the sound eigenvectors[15]

$$\mathbf{a}^{\mathrm{R}}_{\sigma,\mathbf{q}_{\|}n}(\mathbf{r}) = \begin{pmatrix} c\rho^{1/2} p_{\sigma,q_{\|}n}(z) \\ \dfrac{\gamma-1}{\alpha c \rho^{1/2}}\left[p_{\sigma,q_{\|}n}(z) + \left(\dfrac{\gamma}{\gamma-1}\right)^{1/2} S_{\sigma,q_{\|}n}(z)\right] \\ \dfrac{1}{\rho^{1/2} q_{\|}} \nabla\phi_{\sigma,q_{\|}n}(z) \end{pmatrix} \exp(i\mathbf{q}_{\|}\cdot\mathbf{r}_{\|}) \tag{2.12a}$$

$$\mathbf{a}^{\mathrm{L}}_{\sigma,\mathbf{q}_{\|}n}(\mathbf{r}) = \begin{pmatrix} \dfrac{1}{c\rho^{1/2}}\{p_{\sigma,q_{\|}n}(z) - [\gamma(\gamma-1)]^{1/2} S_{\sigma,q_{\|}n}(z)\} \\ \alpha c \rho^{1/2}\left(\dfrac{\gamma}{\gamma-1}\right)^{1/2} S_{\sigma,q_{\|}n}(z) \\ -\dfrac{\rho^{1/2}}{q_{\|}} \nabla\phi_{\sigma,q_{\|}n}(z) \end{pmatrix} \exp(-i\mathbf{q}_{\|}\cdot\mathbf{r}_{\|}) \tag{2.12b}$$

where the scalar functions $p_{\sigma,q_{\|}n}(z)$, $S_{\sigma,q_{\|}n}(z)$, and $\phi_{\sigma,q_{\|}n}(z)$ (depending on $q_{\|} = |\mathbf{q}_{\|}|$ only, due to rotational invariance in the x, y plane) are the eigensolutions of a one-dimensional, 3×3 eigenvalue problem to be given explicitly in the next section. Physically, the three scalar functions correspond to small deviations of the pressure, the entropy density,[3] and the potential of the longitudinal velocity, respectively, from the stationary state. In (2.12), we have not explicitly indicated the dependence of the average quantities on z.

Applying now (2.11) and (2.12) together with the orthogonality and completeness relations satisfied by the eigenvectors to Eqs. (2.6)–(2.8) and (2.10), one obtains a formal expansion of the fast part of the correlation matrix in terms of the sound modes. From this one finds the fast part of the density–density correlation function, which has to be inserted into (2.3) and (2.4) to yield the mode expansion of the Brillouin lines. In doing so, we will further assume that the linear dimension of the scattering volume L_s is intermediate between the wavelength k^{-1} and the macroscopic lengths d and L_∇. In this case, the ratios L_s/L_∇, L_s/d, and $1/L_s k$ are all small,

[3] The entropy density has been identified in Refs. 1 and 2 to be a slow variable in the bulk fluid. Hence, it does not contribute to the sound modes there. In a finite system with given boundary conditions [cf. Eq. (3.2) below], however, the entropy density does not vanish for the sound modes, although it is nonzero only within hydrodynamic boundary layers with thickness of the order $\varepsilon^{1/2}/k$. Similar boundary layer contributions arise in the velocity field. We prove in Appendix B that such boundary layer contributions are negligible for the Brillouin lines as long as the scattering volume is located away from the boundaries.

typically of the order $\sqrt{\varepsilon}$. Neglecting such small corrections, the mode expansion of the Brillouin lines reads[2,16]

$$S_{\mathrm{B}\sigma}(\mathbf{k}, \omega; R_z) = k_{\mathrm{B}} \sum_n \frac{B_{n\sigma}(\mathbf{k}; R_z)}{s_{\sigma,k_\| n} - i\omega} + \text{c.c.} \qquad (\sigma = \pm) \tag{2.13}$$

where the $B_{n\sigma}(\mathbf{k}; R_z)$ are oscillator strengths. Corresponding to the two terms on the rhs of Eq. (2.7), they can be decomposed into a local equilibrium and a mode-coupling part:

$$B_{n\sigma}(\mathbf{k}; R_z) = B_{n\sigma}^{\mathrm{LE}}(\mathbf{k}; R_z) + B_{n\sigma}^{\mathrm{MC}}(\mathbf{k}; R_z) \tag{2.14}$$

These are given by

$$B_{n\sigma}^{\mathrm{LE}}(\mathbf{k}; R_z) = \frac{\rho(R_z)\, T(R_z)}{c^2(R_z)} (2\pi)^2\, \hat{p}_{\sigma,k_\| n}(k_z)\, \hat{p}_{\sigma,k_\| n}(-k_z) \tag{2.15}$$

and

$$B_{n\sigma}^{\mathrm{MC}}(\mathbf{k}, R_z) = -\frac{\rho(R_z)}{c^2(R_z)} (2\pi)^4\, \hat{p}_{\sigma,k_\| n}(k_z) \times \sum_m \frac{\Pi_{\sigma n, -\sigma m}(k_\|)}{s_{\sigma,k_\| n} + s_{-\sigma,k_\| m}}\, \hat{p}_{\sigma,k_\| m} \tag{2.16}$$

where

$$\hat{p}_{\sigma,k_\| n}(k_z) = \frac{1}{L_s^{1/2}} \int_{R_z - L_s/2}^{R_s + L_s/2} e^{-ik_z z} p_{\sigma,k_\| n}(z) \tag{2.17}$$

Finally,

$$\begin{aligned} \Pi_{\sigma n, -\sigma m}(k_\|) = -\frac{1}{k_\|} \int_{-d/2}^{d/2} c(z) \frac{dT}{dz} \Bigg\{ \Bigg[& p_{\sigma,k_\| n}(z) \\ & + \alpha T \left(\frac{\gamma}{\gamma - 1}\right)^{1/2} S_{\sigma,k_\| n}(z) \Bigg] \frac{d\phi_{-\sigma,k_\| m}}{dz} \\ & + \frac{d\phi_{\sigma,k_\| n}}{dz} \left[p_{-\sigma,k_\| m}(z) + \alpha T \left(\frac{\gamma}{\gamma - 1}\right)^{1/2} S_{-\sigma,k_\| m}(z) \right] \Bigg\}\, dz \end{aligned} \tag{2.18}$$

are the matrix elements of the mode-coupling matrix.

Notice that $B_{n\sigma}^{\mathrm{LE}}$ is determined by the mode $(\sigma, \mathbf{k}_\| n)$ alone, while in $B_{n\sigma}^{\mathrm{MC}}$ all modes $(-\sigma, \mathbf{k}_\| m)$ are coupled to $(\sigma, \mathbf{k}_\| n)$ via the mode-coupling matrix.[4] Equations (2.13)–(2.18) are the basis for our further calculations.

[4] In principal, other (slow) hydrodynamic modes couple to $(\sigma, \mathbf{k}_\| n)$ as well. The couplings kept in (2.16) are those for which the denominator becomes small when the leading contributions to the eigenvalues cancel. The error made in neglecting all the other mode couplings is of the order ε.

3. NONEQUILIBRIUM SOUND MODES

In order to proceed with the evaluation of the Brillouin lines, we need the eigenvalues $s_{\sigma,q_{\|}n}$ and eigenfunctions $p_{\sigma,q_{\|}n}(z)$, $S_{\sigma,q_{\|}n}(z)$, and $\phi_{\sigma,q_{\|}n}(z)$. They follow from the eigenvalue problem[15]

$$\begin{aligned} D_T\mathscr{D}S(z)+\left(\frac{\gamma-1}{\gamma}\right)^{1/2} D_T\mathscr{D}p(z)&=sS(z)\\ [\gamma(\gamma-1)]^{1/2} D_T\mathscr{D}S(z)+(\gamma-1)\,D_T\mathscr{D}p(z)-\frac{1}{q_{\|}}\,c\mathscr{D}\phi(z)&=sp(z)\\ q_{\|}\mathscr{D}cp(z)+\Gamma_l\mathscr{D}^2\phi(z)&=s\mathscr{D}\phi(z) \end{aligned} \tag{3.1}$$

where $\mathscr{D}=q_{\|}^2-d^2/dz^2$, and the boundary conditions are

$$\begin{aligned} S\left(\pm\frac{d}{2}\right)+\left(\frac{\gamma-1}{\gamma}\right)^{1/2} p\left(\pm\frac{d}{2}\right)&=0\\ \phi\left(\pm\frac{d}{2}\right)&=0\\ \frac{d\phi}{dz}\left(\pm\frac{d}{2}\right)\mp\beta^{(\pm)}q_{\|}p\left(\pm\frac{d}{2}\right)&=0 \end{aligned} \tag{3.2}$$

In (3.1), the entropy density $S(z)$ is supposed to vanish away from the boundaries. Furthermore, $D_T=\lambda/\rho c_p$ is the thermal diffusivity and $\Gamma_l=(\frac{4}{3}\eta+\zeta)/\rho$ is the longitudinal viscosity, with η and ζ denoting the shear and bulk viscosity, respectively. The z dependence of the average quantities has not been indicated explicitly in (3.1) and (3.2). The first two boundary conditions in (3.2) are the conditions of perfectly heat conducting and sticking plates. The last condition accounts for the sound absorption by the walls. Here $\beta^{(+)}$ and $\beta^{(-)}$ are the specific acoustic admittances[20] of the plates at $z=+d/2$ and $z=-d/2$, respectively.[5] These parameters describe the local distortions of the walls in response to pressure perturbations.[20] In general, they are complex quantities with positive real parts. Special cases are $\beta=0$ (total reflection) and $\beta=1$ (total absorption). The normalization of the eigenfunctions is[15]

$$\int_{-d/2}^{d/2}\left[p^2(z)+\gamma(z)\,S^2(z)-\frac{1}{q_{\|}^2}\,\phi(z)\,\mathscr{D}\phi(z)\right]dz=\frac{1}{(2\pi)^2} \tag{3.3}$$

[5] Since β will in general be temperature dependent, we do not assume $\beta^{(+)}=\beta^{(-)}$.

Making use of the small parameters ε, one can solve the eigenvalue problem (3.1)–(3.3) analytically. Details of the solution will be reported elsewhere.[15] Just two technical points should be mentioned here. First, it is appropriate to use WKB techniques, since the wavelengths of the modes considered here are much smaller than the length on which the macroscopic fields vary [cf. ε_2 in (2.9)]. Second, one must apply a singular perturbation theory in order to take proper care of the hydrodynamic boundary layers close to the plates. We now summarize the results for the nonequilibrium sound modes, as they are relevant for the following, and discuss their physical properties.

The WKB method shows that nonequilibrium sound does not propagate along straight lines, since the sound velocity $c(z)$ is not constant. Instead, the sound rays are curves. A particular sound ray is uniquely determined by fixing a reference point $\mathbf{R}$ in the bulk fluid[6] through which it passes and a wave vector $\mathbf{q}(\mathbf{R}) = (\mathbf{q}_{\|}, q_z(\mathbf{R}))$ in that point. In fact, given $(\mathbf{R}, \mathbf{q}(\mathbf{R}))$ as "initial conditions," the whole ray can be constructed via the tangent vectors $\hat{\mathbf{q}}(z; \mathbf{R}, \mathbf{q}(\mathbf{R}))$, which are the unit vectors corresponding to the local wave vectors

$$\mathbf{q}(z; \mathbf{R}, \mathbf{q}(\mathbf{R})) = (\mathbf{q}_{\|}, q_z(z; \mathbf{R}, \mathbf{q}(\mathbf{R})) \tag{3.4}$$

where

$$q_z(\mathbf{z}; \mathbf{R}, \mathbf{q}(\mathbf{R})) = q_z(\mathbf{R}) \frac{c(R_z)}{c(z)} \left[1 + \frac{c^2(R_z) - c^2(z)}{c^2(R_z)} \frac{q_{\|}^2}{q_z^2(\mathbf{R})} \right]^{1/2} \tag{3.5}$$

In this paper, we assume that the sound rays causing the light scattering are monotonically bent between the plates confining the fluid, i.e., we assume that $q_z(z; \mathbf{R}, \mathbf{q}(\mathbf{R}))$ is nonzero for all z. The case that the sound rays are totally bent will be discussed in another paper.[17] Moreover, we will keep the reference point fixed and suppress the dependence on $\mathbf{R}$, thus writing simply $\mathbf{q}(z; \mathbf{R}, \mathbf{q}(\mathbf{R})) = \mathbf{q}(z, \mathbf{q}(\mathbf{R}))$. By construction, $\mathbf{q}(R_z, \mathbf{q}(\mathbf{R})) = \mathbf{q}(\mathbf{R})$, and the unit vector $\hat{\mathbf{q}}(z, \mathbf{q}(\mathbf{R}))$ defines the local direction of the ray in z. Equations (3.4) and (3.5) are equivalent to

$$c(z)\, q(z, \mathbf{q}(\mathbf{R})) = c(R_z)\, q(\mathbf{R}) \tag{3.6}$$

which is just Snell's law in a stratified medium.[21]

For each $\mathbf{q}_{\|}$ there is only a discrete set of positive[7] values for $q_z(\mathbf{R})$ that allow the boundary conditions to be satisfied at $z = \pm d/2$. We will

[6] We will later choose $\mathbf{R}$ to be center of the scattering volume.

[7] For a complete system of eigenvectors, only modes with $q_z(\mathbf{R}) > 0$ are needed.[15]

denote these characteristic values by q_{nz}, where n is a positive integer.[8] Putting $\mathbf{q}_n = (\mathbf{q}_{\|}, q_{nz})$, the q_{nz} are given by[15]

$$\int_{-d/2}^{d/2} q_z(z, \mathbf{q}_n)\, dz = n\pi \tag{3.7}$$

Only the special rays $(\mathbf{R}, \mathbf{q}_n)$ with $\mathbf{q}_n$ satisfying (3.7) are suitable for eigenmodes.

To define the eigenvalues and eigenfunctions, we need next the effective sound velocity $c(\mathbf{R}, \mathbf{q}_n)$ and the effective sound damping coefficient $\Gamma_s(\mathbf{R}, \mathbf{q}_n)$ associated with the ray $(\mathbf{R}, \mathbf{q}_n)$. These quantities are defined by

$$\frac{1}{c(\mathbf{R}, \mathbf{q}_n)} \frac{d}{\hat{q}_{nz}} = \int_{-d/2}^{d/2} \frac{1}{c(z)} \frac{dz}{\hat{q}_z(z, \mathbf{q}_n)} \tag{3.8}$$

and

$$\frac{\Gamma_s(\mathbf{R}, \mathbf{q}_n)\, q_n^2}{2c(\mathbf{R}, \mathbf{q}_n)} \frac{d}{\hat{q}_{nz}} = \int_{-d/2}^{d/2} \frac{\Gamma_s(z)\, q^2(z, \mathbf{q}_n)}{2c(z)} \frac{dz}{\hat{q}_z(z, \mathbf{q}_n)} \tag{3.9}$$

where $\Gamma_s = \Gamma_l + (\gamma - 1) D_T$ is the local sound damping coefficient. The right-hand side of Eq. (3.8) is the time it takes the ray $(\mathbf{R}, \mathbf{q}_n)$ to pass from plate to plate, while the right-hand side of (3.9) is the total attenuation the ray experiences on that way through the fluid. Thus, $c(\mathbf{R}, q_n)$ and $\Gamma_s(\mathbf{R}, \mathbf{q}_n)$ are averages taken over the ray $(\mathbf{R}, \mathbf{q}_n)$.

Besides the bulk damping, the ray suffers an additional damping caused by the sound absorption of the walls. This depends on the acoustic admittances $\beta^{(+)}$ and $\beta^{(-)}$ of the plates and on the mode considered. Putting[9]

$$b_\sigma(\hat{\mathbf{q}}, \beta) = \frac{\sigma}{2} \ln \frac{\hat{q}_z - i\hat{q}_{\|} + \sigma\beta}{\hat{q}_z + i\hat{q}_{\|} - \sigma\beta} \tag{3.10}$$

with $\hat{q}_{\|} = q_{\|}/q$, we obtain the total surface absorption of the mode $(\sigma, \mathbf{q}_n)$ from[15]

$$\alpha_\sigma(\hat{\mathbf{q}}_n) = b_\sigma^{(+)}(\hat{\mathbf{q}}_n) + b_\sigma^{(-)}(\hat{\mathbf{q}}_n) \tag{3.11}$$

where

$$b_\sigma^{(\pm)}(\hat{\mathbf{q}}_n) = b_\sigma(\hat{\mathbf{q}}(\pm d/2, \mathbf{q}_n), \beta^{(\pm)}) \tag{3.12}$$

[8] The integer n is large for the modes considered here, typically of the order ε^{-1}. For small n our WKB method is not applicable.

[9] In (3.10), it is understood that the main branch of the logarithm is taken (see Ref. 22).

In order to be able to write the nonequilibrium sound modes in a compact form, it is convenient to introduce the complex, local wave vectors

$$\mathbf{Q}_\sigma(z, \mathbf{q}_n) = (\mathbf{q}_\|, Q_{\sigma z}(z, \mathbf{q}_n)) \tag{3.13}$$

the z components of which are given by

$$Q_{\sigma z}(z, \mathbf{q}_n) = q_z(z, \mathbf{q}_n) + i\eta_\sigma(z, \mathbf{q}_n) \tag{3.14}$$

where $\eta_\sigma(z, \mathbf{q}_n)$ is a small correction term of the order ε relative to $q_z(z, \mathbf{q}_n)$, which is defined by

$$\eta_\sigma(z, \mathbf{q}_n) = \frac{\sigma}{2c(z)\,\hat{q}_z(z, \mathbf{q}_n)}\,[\Gamma_s(z)\,q^2(z, \mathbf{q}_n) - \Gamma_s(\mathbf{R}, \mathbf{q}_n)\,q_n^2]$$
$$-\frac{\sigma}{d}\,\frac{c(\mathbf{R}, \mathbf{q}_n)\,\hat{q}_{nz}}{c(z)\,\hat{q}_z(z, \mathbf{q}_n)}\,\alpha_\sigma(\hat{\mathbf{q}}_n) \tag{3.15}$$

The real part of $\eta_\sigma(z, \mathbf{q}_n)$ consists of a surface term, proportional to $(1/d)\,\mathrm{Re}\,\alpha_\sigma(\hat{\mathbf{q}}_n) > 0$, and that describes the sound absorption by the two plates, and a bulk term. The latter gives rise to a spatial envelope that enhances the attenuation length in the direction of decreasing $\Gamma_s(z)$, and diminishes it in the direction of increasing $\Gamma_s(z)$.[15] This is a pure non-equilibrium term. From (3.7)–(3.9) and (3.15) it follows that

$$\int_{-d/2}^{d/2} Q_{\sigma z}(z, \mathbf{q}_n)\,dz = n\pi - i\sigma\alpha_\sigma(\hat{\mathbf{q}}_n) \tag{3.16}$$

After these definitions we are now in a position to present the eigenmodes. Up to first order in ε the eigenvalues read[15]

$$s_{\sigma, q_\| n} = i\sigma c(R_z)\,Q_\sigma(R_z, \mathbf{q}_n) + \frac{1}{2}\,\Gamma_s(R_z)\,Q_\sigma^2(R_z, \mathbf{q}_n) \tag{3.17}$$

We remark that the complex wave vectors have been constructed in such a way that the eigenvalues do not depend on the reference point (up to first order in ε), as it should be. From the three zeroth-order eigenfunctions we give here only the pressure eigenfunctions, because only they enter directly in the expressions (2.14)–(2.17) for the oscillator strengths. The pressure eigenfunctions are

$$p_{\sigma, q_\| n}(z) = \frac{1}{2\pi\sqrt{d}}\left[\frac{c(\mathbf{R}, q_n)\,\hat{q}_{nz}}{c(z)\,\hat{q}_z(z, \mathbf{q}_n)}\right]^{1/2}$$
$$\times \cos\left[\int_{-d/2}^{z} Q_{\sigma z}(z', \mathbf{q}_n)\,dz' + i\sigma b_\sigma^{(-)}(\mathbf{q}_n)\right] \tag{3.18}$$

The other eigenfunctions $S_{\sigma,q_{\|}n}(z)$ and $\phi_{\sigma,q_{\|}n}(z)$ are only needed in the computation of the mode-coupling matrix [cf. Eq. (2.18)], so that we can postpone them to Appendix B.

4. OSCILLATOR STRENGHTS

The nonequilibrium sound modes presented in the last section can now be inserted into the formal mode expansion of the Brillouin lines, Eqs. (2.13)–(2.18). In doing so, it is convenient to identify the reference point **R** with the center of the scattering volume. The remaining problem is then the summation of all the modes. Making use of the small parameter ε, we can carry out all the sums analytically. In this section, we sketch the first step: the evaluation of the oscillator strengths $B_{n\sigma}^{\mathrm{LE}}$ and $B_{n\sigma}^{\mathrm{MC}}$ defined in (2.15) and (2.16), respectively.

First we need the Fourier transforms, defined in (2.17), of the pressure eigenfunctions (3.18). Recalling from Section 2 that L_s is intermediate between the wavelength and the macroscopic lengths, one finds that $\hat{p}_{\sigma,k_{\|}n}(k_z)$, considered as a function of q_{nz}, is concentrated upon a peak of height $\sim L_s$ and width $\sim L_s^{-1}$ around $q_{nz} \approx |k_z|$. Neglecting terms of the order $\sqrt{\varepsilon}$, we obtain

$$\hat{p}_{\sigma,k_{\|}n}(k_z) = \frac{1}{2\pi\sqrt{d}} \left[\frac{c(\mathbf{R}, k)}{c(R_z)}\right]^{1/2} [\exp(-ik_z R_z)]\, \pi_\sigma(q_{nz}, k_z)$$
$$\times \exp i \frac{k_z}{|k_z|} \left[\int_{-d/2}^{R_z} Q_{\sigma z}(z, \mathbf{q}_n)\, dz + i\sigma b_n^{(-)}(\hat{\mathbf{q}}_n)\right] \qquad (4.1)$$

where

$$\pi_\sigma(q_{nz}, k_z) = \frac{1}{2\sqrt{L_s}} \int_{-L_s/2}^{L_s/2} dz \exp i \left[(q_{nz} + i\bar{\eta}_\sigma - |k_z|)\, z - \frac{1}{2\hat{k}_z} \frac{d \ln c}{dR_z} k z^2\right] \qquad (4.2)$$

and we have put $\mathbf{q}_n = (\mathbf{k}_{\|}, q_{nz})$ and $\bar{\eta}_\sigma = \eta_\sigma(R_z, \mathbf{k})$. Inserting (4.1) and (4.2) into (2.15), we find for the local equilibrium oscillator strengths, after some straightforward manipulations summarized in Appendix A,

$$B_{n\sigma}^{\mathrm{LE}}(\mathbf{k}; R_z) = \frac{\rho(R_z)\, T(R_z)}{2c^2(R_z)} \frac{\pi c(\mathbf{R}, \mathbf{k})}{dc(R_z)} \Delta_1(q_{nz} + i\bar{\eta}_\sigma - |k_z|) \qquad (4.3)$$

where

$$\Delta_1(q) = \frac{1}{\pi} \frac{\sin^2(q L_s/2)}{q^2 L_s/2} \qquad (4.4)$$

In computing the mode-coupling part of the oscillator strengths, given by (2.16), we again make use of the fact that the functions $\hat{p}_{\sigma,k_{\|}n}(k_z)$ and $\hat{p}_{-\sigma,k_{\|}m}(-k_z)$ are peaked around $q_{nz} \approx |k_z|$ and $q_{mz} \approx |k_z|$, respectively. For this reason we need to take into account in the sum only those terms where q_{mz} lies within a distance of the order L_s^{-1} of q_{nz}. In other words, we have to take into account only modes with values m close to n, i.e., $|m-n|/n \leqslant O(\sqrt{\varepsilon})$. Putting thus $\mathbf{q}_m = (\mathbf{k}_{\|}, q_{mz})$ and expanding around q_{nz}, we obtain from (3.5)–(3.8) for the modes in question

$$q_z(z, \mathbf{q}_m) = q_z(z, \mathbf{q}_n) + (m-n)\frac{\pi}{d}\frac{c(\mathbf{R}, \mathbf{k})\,|\hat{k}_z|}{c(z)\,|\hat{q}_z(z, \mathbf{k})|} \tag{4.5}$$

where

$$q_z(z, \mathbf{k}) = k_z \frac{c(R_z)}{c(z)}\left[1 + \frac{c^2(R_z) - c^2(z)}{c^2(R_z)}\frac{k_{\|}^2}{k_z^2}\right]^{1/2} \tag{4.6}$$

In the second term on the right-hand side of (4.5), we have neglected terms of the order $\sqrt{\varepsilon}$ in replacing q_{nz} by $|\hat{k}_z|$. Similar to (4.5), we can evaluate $\Pi_{\sigma n, -\sigma m}(k_{\|})$, $\hat{p}_{-\sigma,k_{\|}m}(-\hat{k}_z)$, and $s_{-\sigma,k_{\|}m}$ for values m close to n. In this way, we obtain the leading approximations to all the quantities we have to sum in (2.16). These results will be given next.

For the matrix elements of the mode-coupling matrix we find

$$\Pi_{\sigma n, -\sigma m}(k_{\|}) = \frac{i\sigma}{(2\pi)^2 d} c(\mathbf{R}, \mathbf{k})\,|\hat{k}_z| \times \int_{-d/2}^{d/2} dz \frac{dT}{dz} \sin[(m-n)\,\pi\chi_1(z) - i\sigma\chi_2(z)] \tag{4.7}$$

where we have introduced the auxiliary functions

$$\chi_1(z) = \chi_1(z, \mathbf{k}) = c(\mathbf{R}, \mathbf{k})\,|\hat{k}_z|\,\frac{1}{d}\int_{-d/2}^{z} \frac{1}{c(z')}\frac{dz'}{|\hat{q}_z(z', \mathbf{k})|} \tag{4.8}$$

and

$$\chi_2(z) = \chi_2(z, \mathbf{k}) = \int_{-d/2}^{z} \frac{\Gamma_s(z')\,q^2(z', \mathbf{k}) - \Gamma_s(\mathbf{R}, \mathbf{k})\,k^2}{c(z')}\frac{dz'}{|\hat{q}_z(z', \mathbf{k})|} - 2\chi_1(z)\,w^{(+)}(\hat{\mathbf{k}}) + 2[1-\chi_1(z)]\,w^{(-)}(\hat{\mathbf{k}}) \tag{4.9}$$

In (4.9), we have used the total absorption coefficients $w^{(+)}(\hat{\mathbf{k}})$ and $w^{(-)}(\hat{\mathbf{k}})$ of the walls at $z = +d/2$ and $z = -d/2$, respectively. They are defined by

$$w^{(\pm)}(\hat{\mathbf{k}}) = \frac{1}{2}\,[b_\sigma^{(\pm)}(\hat{\mathbf{k}}) + b_{-\sigma}^{(\pm)}(\hat{\mathbf{k}})] \tag{4.10}$$

More details on the derivation of (4.7)–(4.10) are given in Appendix B.

From (3.11), (3.14), (3.15), (4.1), and (4.5) we find, furthermore,

$$\hat{p}_{\sigma,k_\parallel n}(k_z)\,\hat{p}_{-\sigma,k_\parallel m}(-k_z) = \frac{1}{(2\pi)^2 d}\frac{c(\mathbf{R},\mathbf{k})}{c(R_z)}\,\pi_\sigma(q_{nz},k_z)\,\pi_{-\sigma}(q_{mz},-k_z) \times \exp -i\frac{k_z}{|k_z|}\left[(m-n)\,\pi\chi_1(R_z)-i\sigma\chi_2(R_z)\right] \tag{4.11}$$

and, using (3.11), (3.13)–(3.15), (3.17), and (4.5), we obtain

$$s_{\sigma,k_\parallel n}+s_{-\sigma,k_\parallel m} = -i\sigma\frac{\pi}{d}c(\mathbf{R},\mathbf{k})\,|\hat{k}_z|\left[(m-n)+i\sigma\frac{2}{\pi}\xi(\mathbf{k})\right] \tag{4.12}$$

where

$$\xi(\mathbf{k})=\frac{\Gamma_s(\mathbf{R},\mathbf{k})\,k^2}{2c(\mathbf{R},\mathbf{k})}\frac{d}{|\hat{k}_z|}+w^{(+)}(\hat{\mathbf{k}})+w^{(-)}(\hat{\mathbf{k}}) \tag{4.13}$$

With the aid of the expressions (4.7), (4.11), and (4.12), one can now compute the sum over m in (2.16) in closed form. This calculation is outlined in Appendix C. The result is

$$B_{n\sigma}^{\mathrm{MC}}(\mathbf{k},R_z)=\frac{\rho(R_z)}{2c^2(R_z)}\frac{\pi c(\mathbf{R},\mathbf{k})}{dc(R_z)} \times \Delta_\sigma^{\mathrm{MC}}(q_{nz}+i\bar{\eta}_\sigma-|k_z|;\mathbf{k})\int_{-d/2}^{d/2}\frac{dT}{dz}G_\sigma(z;\mathbf{R},\mathbf{k}) \tag{4.14}$$

In (4.14), the quantity

$$\Delta_\sigma^{\mathrm{MC}}(q_{nz}+i\bar{\eta}_\sigma-|k_z|;\mathbf{k})=\frac{2}{\pi}\pi_\sigma(q_{nz},k_z)\,\pi_{-\sigma}(q'_{nz},-k_z) \tag{4.15}$$

with

$$q'_{nz}=q'_{nz}(\mathbf{k})=q_{nz}-i\sigma\frac{2}{d}\frac{c(\mathbf{R},\mathbf{k})}{c(R_z)}\xi(\mathbf{k}) \tag{4.16}$$

will be seen in the next section to give rise to deviations of the line shapes from the Lorentzian form. Thus, we will call $\Delta_\sigma^{\mathrm{MC}}$ the line-shape factor. Furthermore, the dimensionless function $G_\sigma(z;\mathbf{R},\mathbf{k})$ appearing under the integral in (4.14) will be called the propagator. It plays a central role in our theory, since it measures the range over which the temperature gradient is "felt." The propagator is explicitly given by

$$G_\sigma(z;\mathbf{R},\mathbf{k})=\begin{cases}-\dfrac{\exp 2\sigma(k_z/|k_z|)\,\xi^{(+)}(R_z,\mathbf{k})}{\sinh 2\xi(\mathbf{k})}\sinh 2\xi^{(-)}(z,\mathbf{k}) \\ \left(-\dfrac{d}{2}\leqslant z<R_z\right) \\ \dfrac{\exp -2\sigma(k_z/|k_z|)\,\xi^{(-)}(R_z,\mathbf{k})}{\sinh 2\xi(\mathbf{k})}\sinh 2\xi^{(+)}(z,\mathbf{k}) \\ \left(R_z<z\leqslant\dfrac{d}{2}\right)\end{cases} \tag{4.17}$$

where

$$\xi^{(\pm)}(z,\mathbf{k})=\pm\int_z^{\pm d/2}\frac{\Gamma_s(z')\,q^2(z',\mathbf{k})}{2c(z')\,|\hat{q}_z(z',\mathbf{k})|}\,dz'+w^{(\pm)}(\hat{\mathbf{k}}) \tag{4.18}$$

is the total sound attenuation between the point $\mathbf{r}$ on the ray $(\mathbf{R},\mathbf{k})$ and the plate located at $\pm d/2$. It consists of a fluid term and a wall term. Notice from (3.9), (4.13), and (4.18) that

$$\xi^{(+)}(z,\mathbf{k})+\xi^{(-)}(z,\mathbf{k})=\xi(\mathbf{k}) \tag{4.19}$$

for all z.

The last step in the evaluation of the mode-coupling oscillator strengths consists in computing the line-shape factor $\Delta_\sigma^{\mathrm{MC}}$ defined in (4.15) and (4.16). This calculation is summarized in Appendix D, with the result

$$\Delta_\sigma^{\mathrm{MC}}(q,\mathbf{k})=\Delta_1(q)-\sigma\frac{\Gamma_s(R_z)\,k^2}{2c(R_z)|\hat{k}_z|}\times\left[i\frac{\partial\Delta_1(q)}{\partial q}-\frac{1}{6\hat{k}_z}\frac{d\ln c}{dR_z}kL_s^2\frac{\partial\Delta_2(q)}{\partial q}\right] \tag{4.20}$$

where $\Delta_1(q)$ has been defined in (4.4) and

$$\Delta_2(q)=-\frac{3}{2\pi}\frac{\sin^2(qL_s/2)-(qL_s/2)^2}{q^4(L_s/2)^3} \tag{4.21}$$

To close this section, we note that the total sound attenuations $\xi^{(\pm)}(z,\mathbf{k})$ always appear with a factor 2 in (4.17). This is due to mode-coupling: one term can be traced back to the "host" mode $(\sigma,\mathbf{q}_n)$ in (2.16), while the contributions of all the coupled modes $(-\sigma, \mathbf{q}_m)$ in the sum (2.16) add up to the same result. Mode coupling is also the origin of the second term on the right-hand side of (4.20), which will be found to cause non-Lorentzian line shapes.

5. RESULTS AND DISCUSSION

Knowing the eigenvalues (3.17) and the oscillator strengths (4.3) and (4.14), one can return to (2.13) and evaluate, finally, the Brillouin lines in the stationary state. This step of the calculation is outlined in Appendix E. Here, we will present immediately our final results and conclude this paper by some remarks concerning their physical interpretation.

For the local equilibrium part of the Brillouin lines, we obtain

$$S_{B\sigma}^{LE}(\mathbf{k}, \omega; R_z) = I_B(R_z) \operatorname{Re} Y_{B\sigma}(\mathbf{k}, \omega; R_z) \tag{5.1}$$

where

$$I_B(R_z) \equiv \frac{1}{2\pi} \int_{-\infty}^{\infty} S_{B\sigma}^{LE}(\mathbf{k}, \omega; R_z)\, d\omega = k_B T(R_z) \frac{\rho(R_z)}{2c^2(R_z)} \tag{5.2}$$

is the local equilibrium part of the integrated intensity of each line, while

$$Y_{B\sigma}(\mathbf{k}, \omega; R_z) = \frac{2}{-i[\omega - \sigma c(R_z)\, k] + \frac{1}{2}\Gamma_s(R_z)\, k^2} \tag{5.3}$$

describes the line shape. The local equilibrium intensity $I_B(R_z)$ does not depend on $\mathbf{k}$ and is the same for both lines ($\sigma = +$ and $\sigma = -$); the local equilibrium line shapes are Lorentzian. By Eqs. (5.1)–(5.3), we have verified that our nonequilibrium sound modes lead to the expected answer for the local equilibrium part of the dynamic structure factor (Ref. 18, p. 243). We need not discuss the local equilibrium results further.

The main results of this paper are our expressions for the mode-coupling, i.e., the typical nonequilibrium contributions to the Brillouin lines. They are the following:

$$S_{B\sigma}^{MC}(\mathbf{k}, \omega; R_z) = \operatorname{Re}[I_{B\sigma}^{MC}(\mathbf{k}; R_z) \cdot Y_{B\sigma}^{MC}(\mathbf{k}, \omega; R_z)] \tag{5.4}$$

In (5.4) the mode-coupling intensities read

$$I_{B\sigma}^{MC}(\mathbf{k}; R_z) = I_B(R_z) \frac{1}{T(R_z)} \int_{-d/2}^{d/2} \frac{dT}{dz}\, G_\sigma(z; \mathbf{R}, \mathbf{k}) \tag{5.5}$$

with $G_\sigma(z; \mathbf{R}, \mathbf{k})$ the propagator on the ray $(\mathbf{R}, \mathbf{k})$ as defined in (4.17) and (4.18). Furthermore, the line shapes in (5.4) are given by

$$Y_{B\sigma}^{MC}(\mathbf{k}, \omega; R_z) = \left[1 - \Omega(\mathbf{k}; R_z)\frac{\partial}{\partial\omega}\right] Y_{B\sigma}(\mathbf{k}, \omega; R_z) \tag{5.6}$$

where

$$\Omega(\mathbf{k}; R_z) = \frac{1}{2}\Gamma_s(R_z)\, k^2 \left(i - \frac{1}{6\hat{k}_z}\frac{d \ln c}{dR_z} k L_s^2\right) \tag{5.7}$$

The expressions (5.4)–(5.7) are new. They take into account simultaneously both the boundary effects (partially or totally sound-absorbing walls) and the nonlinear effects (spatial inhomogeneities induced by the temperature gradient). The mode-coupling intensity $I_{B\sigma}^{MC}(\mathbf{k}; R_z)$ given by (5.5) is proportional to the temperature difference between the lower and the upper plate, weighted by the propagator $G_\sigma(z; \mathbf{R}, \mathbf{k})$ on the particular ray $(\mathbf{R}, \mathbf{k})$ that is selected by the scattering geometry.

We conclude this paper with a few remarks on the expressions (5.4)–(5.7). A more detailed discussion, involving the relationship of the present theory with those of Satten and Ronis[10] and that of Kirkpatrick *et al.*,[14] as well as new experimental checks of the theory, will be reserved for another paper.[17]

1. The linear theory for infinite systems[4–9] is, of course, contained in our theory as the simplest special case. In fact, neglecting the spatial inhomogeneities in putting $c(z) = c(R_z)$ and $\Gamma_s(z) = \Gamma_s(R_z)$ and taking the limit $d \to \infty$, one obtains from (4.6), (4.13), and (4.17)–(4.19) for the propagators

$$G_\sigma(z; \mathbf{R}, \mathbf{k}) = -\sigma\frac{k_z}{|k_z|}\,\Theta\left(\sigma\frac{k_z}{|k_z|}(R_z - z)\right) \times \exp -2\sigma\frac{\Gamma_s(R_z)\, k^2}{2c(R_z)}\frac{1}{\hat{k}_z}(R_z - z) \tag{5.8}$$

where $\Theta(z)$ is the Heaviside step function. Inserting this into (5.5) with $dT/dz = dT/dR_z$, $d \to \infty$, yields

$$I_{B\sigma}^{MC}(\mathbf{k}; R_z) = -I_B(R_z)\frac{\sigma}{T(R_z)}\frac{dT}{dR_z}\hat{k}_z\frac{c(R_z)}{\Gamma_s(R_z)\, k^2} \tag{5.9}$$

which is the well-known linear result. Equation (5.9) implies that the scattering intensity is higher (lower) for the Brillouin line that probes the sound wave coming from the warmer (cooler) side of the system by an amount proportional to the temperature difference "felt" by the (straight) sound ray $(\mathbf{R}, \mathbf{k})$ along half[10] a mean free path[(24)]

$$l_{\mathrm{mfp}}(k) = 2c(R_z)/\Gamma_s(R_z)\, k^2$$

The linear attenuation length $l_{\mathrm{mfp}}(k)$ enters in the exponent of our propagator (5.8).

2. The modifications caused by boundaries and nonlinearities can be seen most immediately in the case of totally absorbing walls, i.e., $\beta^{(+)} = \beta^{(-)} = 1$, and vertical scattering, i.e., $|\hat{k}_z| = 1$. Here one finds from (3.10), (3.12), (4.6), (4.10), (4.13), and (4.17)–(4.19)

$$G_\sigma(z; \mathbf{R}, \mathbf{k}) = -\sigma \frac{k_z}{|k_z|}\, \Theta\left(\sigma \frac{k_z}{|k_z|}(R_z - z)\right) \times \exp -2\sigma \frac{k_z}{|k_z|} \int_z^{R_z} \frac{\Gamma_s(z')\, q^2(z', \mathbf{k})}{2c(z')} \frac{dz'}{|\hat{q}_z(z', \mathbf{k})|} \tag{5.10}$$

This is the generalization of (5.8) to the nonlinear regime, the linear attenuation in the exponent being replaced by the corresponding quantity along the curved sound ray $(\mathbf{R}, \mathbf{k})$. Inserting (5.10) into (5.5), one finds

$$I_{\mathrm{B}\sigma}^{\mathrm{MC}}(\mathbf{k}; R_z) = I_{\mathrm{B}}(R_z) \frac{1}{T(R_z)} \int_{R_z}^{-\sigma(k_z/|k_z|)d/2} dz \frac{dT}{dz} \times \exp -2\sigma \frac{k_z}{|k_z|} \int_z^{R_z} \frac{\Gamma_s(z')\, q^2(z', \mathbf{k})}{2c(z')} \frac{dz'}{|q_z(z', \mathbf{k})|} \tag{5.11}$$

As a result of the spatial inhomogeneities, the attenuation length for the sound propagating in the direction of increasing (decreasing) $\Gamma_s(z)$ is smaller (larger) than the value $l_{\mathrm{mfp}}(k)$ predicted by the linear theory. This purely nonlinear effect can be probed in measuring the sum $(I_{\mathrm{B}+}^{\mathrm{MC}} + I_{B-}^{\mathrm{MC}})$ of the mode-coupling intensities.[(17)] If $\beta \neq 1$, one must use the more complicated expressions (4.17) and (4.18) for the propagators, which take into account the reflection of sound at the walls.

3. The shape of the mode-coupling contribution to the Brillouin lines, given by (5.6), consists of a Lorentzian and a non-Lorentzian part, which is proportional to the derivative of a Lorentzian. The strength of this non-Lorenzian part is determined by the factor $\Omega(\mathbf{k}; R_z)$, defined in (5.7). This factor consists of two terms. The first term on the right-hand side of

[10] The factor 1/2 is due to mode coupling (cf. the comment at the end of the last section).

(5.7) describes a narrowing of the lines and has already been obtained in the linear theories.[4–9] The second term only arises in a nonlinear theory and accounts for a shift in the location of the peaks due to the variation of the sound velocity within the scattering volume. The precise amount of this shift depends very much on the spatial distribution of the intensity of the incoming laser beam. In this paper, we have made the simplest possible choice in assuming a step function with width L_s. Probably a Gaussian distribution, as used in Ref. 10, would have been more realistic. However, since with present techniques the line shapes are almost impossible to measure, it was not our aim to present here quantitative results on the line shift, but merely to give the qualitative trend of the effect. Thus, we obtain from (5.6) and (5.7) to first order in the dimensionless parameter $[(d \ln c)/dR_z]\, kL_s^2$ that the maxima of the lines are located at

$$\omega_\sigma(\mathbf{k}) = \sigma c(R_z)\, k - \frac{1}{48\hat{k}_z} \Gamma_s k^2 \frac{d \ln c}{dR_z} kL_s^2 \tag{5.12}$$

i.e., the sign of the shift is given by the sign of $(dc/dR_z)\, k_z$. Moreover, the lines are no longer symmetrical about their maxima.

APPENDIX A

In this Appendix, we derive the expressions (4.3) and (4.4) for the local equilibrium oscillator strengths. Inserting (4.1) and (4.2) into (2.15) yields

$$\begin{aligned} B_{n\sigma}^{\mathrm{LE}}(\mathbf{k}; R_z) = {} & \frac{\rho(R_z)\, T(R_z)}{c^2(R_z)} \frac{1}{d} \frac{c(\mathbf{R}, \mathbf{k})}{c(R_z)} \\ & \times \frac{1}{4L_s} \int_{-L_s/2}^{L_s/2} dz_1 \int_{-L_s/2}^{L_s/2} dz_2 \\ & \times \exp i \Big[(q_{nz} + i\bar{\eta}_\sigma - |k_z|)(z_1 + z_2) \\ & - \frac{1}{2\hat{k}_z} \frac{d \ln c}{dR_z} k(z_1^2 - z_2^2) \Big] \end{aligned} \tag{A.1}$$

Introducing the new integration variables $\zeta = (1/2L_s)(z_1 + z_2)$ and $z = z_1 - z_2$, one can easily integrate with respect to z, and one finds after a short calculation

$$\begin{aligned} B_{n\sigma}^{\mathrm{LE}}(\mathbf{k}; R_z) = {} & \frac{\rho(R_z)\, T(R_z)}{2c^2(R_z)} \frac{c(\mathbf{R}, \mathbf{k})}{dc(R_z)} \\ & \times \int_0^{1/2} d\zeta \frac{\sin 2(q_{nz} + i\bar{\eta}_\sigma - |k_z|)\, L_s \zeta}{q_{nz} + i\bar{\eta}_\sigma - |k_z|} f\left(\frac{1}{|\hat{k}_z|} \frac{d \ln c}{dR_z} kL_s^2, \zeta \right) \end{aligned} \tag{A.2}$$

where

$$f(a, \zeta) = \frac{d}{d\zeta} \frac{\sin a\zeta(2\zeta - 1)}{a\zeta} \tag{A.3}$$

Since

$$a = \frac{1}{|\hat{k}_z|} \frac{d \ln c}{dR_z} kL_s^2$$

is at most of the order 1, one has to a very good approximation[11]

$$f(a, \zeta) \approx 2 \tag{A.4}$$

in the whole interval $0 \leqslant \zeta \leqslant 1/2$. Using (A.4) in (A.2) and integrating, one finds the results (4.3), (4.4).

APPENDIX B

In this Appendix, we derive the results (4.7)–(4.10) for the elements $\Pi_{\sigma n, -\sigma m}(k_\|)$ of the mode-coupling matrix. According to the definition (2.18), not only are the pressure eigenfunctions $p_{\sigma, q_\| n}(z)$ given in (3.18) required in the calculation, but so are the eigenfunctions $S_{\sigma, q_\| n}(z)$ and $\phi_{\sigma, q_\| n}(z)$, corresponding to the entropy density and the potential of the longitudinal velocity, respectively. They are found in Ref. 15 to read

$$S_{\sigma, q_\| n}(z) = -\frac{1}{2\pi\sqrt{d}} \left[\frac{\gamma(z) - 1}{\gamma(z)} \frac{c(\mathbf{R}, \mathbf{q}_n)\, \hat{q}_{nz}}{c(z)\, \hat{q}_z(z, \mathbf{q}_n)}\right]^{1/2} B^{(2)}_{\sigma, \mathbf{q}_n}(z) \tag{B.1}$$

$$\begin{aligned} \phi_{\sigma, q_\| n}(z) = &-\frac{i\sigma}{2\pi\sqrt{d}} \left[\frac{c(\mathbf{R}, \mathbf{q}_n)\, \hat{q}_{nz}}{c(z)\, \hat{q}_z(z, \mathbf{q}_n)}\right]^{1/2} \hat{q}_\|(z, \mathbf{q}_n) \\ &\times \left\{\cos\left[\int_{-d/2}^{z} Q_{\sigma z}(z', \mathbf{q}_n)\, dz' + i\sigma b_\sigma^{(-)}(\hat{\mathbf{q}}_n)\right] - B^{(1)}_{\sigma, \mathbf{q}_n}(z)\right. \\ &\left. - i\sigma \frac{[\gamma(z) - 1]\, D_T(z)\, q(z, \mathbf{q}_n)}{c(z)} B^{(2)}_{\sigma, \mathbf{q}_n}(z)\right\} \end{aligned} \tag{B.2}$$

Here, $B^{(1)}_{\sigma, \mathbf{q}_n}(z)$ and $B^{(2)}_{\sigma, \mathbf{q}_n}(z)$ are boundary layer functions, defined by

$$B^{(1)}_{\sigma, \mathbf{q}_n}(z) = (-1)^n E_\sigma^{(+)}(\hat{\mathbf{q}}_n)\, e^{q_\|(z - d/2)} + E_\sigma^{(-)}(\hat{\mathbf{q}}_n)\, e^{-q_\|(z + d/2)} \tag{B.3}$$

[11] For example, for $a = 3$ the error is below 5%.

and

$$B^{(2)}_{\sigma,\mathbf{q}_n}(z) = (-1)^n E^{(+)}_\sigma(\hat{\mathbf{q}}_n) \exp\left\{e^{-i\sigma\pi/4}\left[\frac{c(d/2)\,q(d/2,\mathbf{q}_n)}{D_T(d/2)}\right]^{1/2}\left(z-\frac{d}{2}\right)\right\}$$
$$+E^{(-)}_\sigma(\hat{\mathbf{q}}_n)\exp-\left\{e^{-i\sigma\pi/4}\left[\frac{c(-d/2)\,q(-d/2,\mathbf{q}_n)}{D_T(-d/2)}\right]^{1/2}\left(z+\frac{d}{2}\right)\right\} \tag{B.4}$$

where

$$E^{(\pm)}_\sigma(\hat{\mathbf{q}}_n) = \frac{\hat{q}_z(\pm d/2, \mathbf{q}_n)}{[1-\beta^{(\pm)^2}+2i\sigma\beta^{(\pm)}\hat{q}_\parallel(\pm d/2,\mathbf{q}_n)]^{1/2}} \tag{B.5}$$

The boundary layer functions decay exponentially to zero as z is moved away from the walls into the bulk fluid. In fact, Eqs. (B.3) and (B.4) imply that the thickness of the boundary layer corresponding to $B^{(1)}_{\sigma,\mathbf{q}_n}$ is of the order $1/q_n$, while that of $B^{(2)}_{\sigma,\mathbf{q}_n}$ is even only of the order $\varepsilon^{1/2}/q_n$. The amplitudes $E^{(\pm)}_\sigma(\hat{\mathbf{q}}_n)$ depend on the acoustic admittances $\beta^{(\pm)}$ of the plates.

Now we are able to discuss the integral (2.18). Recalling that we need only modes with $q_{nz} \approx |k_z|$ and $q_{mz} \approx |k_z|$, we find from (3.18), (B.1), and (B.2) to leading order in ε for the terms appearing in the integrand of (2.18)

$$p_{\sigma,k_\parallel n}(z) + \alpha T\left(\frac{\gamma}{\gamma-1}\right)^{1/2} S_{\sigma,k_\parallel n}(z)$$
$$= \frac{1}{2\pi\sqrt{d}}\left[\frac{c(\mathbf{R},\mathbf{k})\,|\hat{k}_z|}{c(z)\,|\hat{q}_z(z,\mathbf{k})|}\right]^{1/2}$$
$$\times\left\{\cos\left[\int_{-d/2}^{z} Q_{\sigma z}(z',\mathbf{q}_n)\,dz' + i\sigma b^{(-)}_\sigma(\hat{\mathbf{k}})\right] - \alpha(z)\,T(z)\,B^{(2)}_{\sigma,\mathbf{q}_n}(z)\right\} \tag{B.6}$$

and

$$\frac{d\phi_{-\sigma,k_\parallel m}(z)}{dz}$$
$$= -\frac{i\sigma}{2\pi\sqrt{d}}\left[\frac{c(\mathbf{R},\mathbf{k})\,|\hat{k}_z|}{c(z)\,|\hat{q}_z(z,\mathbf{k})|}\right]^{1/2} k_\parallel\,|\hat{q}_z(z,\mathbf{k})|$$
$$\times\left\{\sin\left[\int_{-d/2}^{z} Q_{-\sigma z}(z',\mathbf{q}_m)\,dz' - i\sigma b^{(-)}_\sigma(\hat{\mathbf{k}})\right]\right.$$
$$\left.+\frac{1}{|q_z(z,\mathbf{k})|}\frac{dB^{(1)}_{-\sigma,\mathbf{q}_m}(z)}{dz}\right\} \tag{B.7}$$

with $\mathbf{q}_n = (\mathbf{k}_\parallel, q_{nz})$, $\mathbf{q}_m = (\mathbf{k}_\parallel, q_{mz})$. In (B.7), we have used (3.14). Furthermore, we have neglected in (B.7) the term containing the derivative of the boundary layer function $B^{(2)}_{-\sigma,\mathbf{q}_m}(z)$. This is justified, although $dB^{(2)}/dz$ is large (i.e., of the order $k/\sqrt{\varepsilon}$), because it is multiplied by a factor of the order ε. Indeed, the function $B^{(2)}_{\sigma,\mathbf{q}_n}$ in (B.2) becomes relevant only when second- or higher order derivatives of $\phi_{\sigma,q_{\parallel n}}(z)$ with respect to z are considered.

When (B.6) and (B.7) are inserted into (2.18), one obtains three types of integrals, which can be estimated as follows:

$$\int_{-d/2}^{d/2} dz\, f(z)\, B^{(2)}_{\sigma,\mathbf{q}_n}(z) = O\left(\frac{\sqrt{\varepsilon}}{k} f\right)$$

$$\times \int_{-d/2}^{d/2} dz\, f(z) \frac{1}{|q_z(z, \mathbf{k})|} \frac{dB^{(1)}_{\sigma,\mathbf{q}_n}(z)}{dz} = O\left(\frac{1}{k} f\right) \tag{B.8}$$

$$\int_{-d/2}^{d/2} dz\, f(z) \cos\left[\int_{-d/2}^{z} Q_{\sigma z}(z', \mathbf{q}_n)\, dz' + i\sigma b^{(-)}_{\sigma}(\hat{\mathbf{k}})\right] = \begin{cases} O(k^{-1} f) & \text{if } f \text{ exponentially decaying} \\ O(fd) & \text{if } f \text{ oscillating} \end{cases}$$

Here $f(z)$ stands for a function that decays exponentially or oscillates on the scale k^{-1}, i.e., $d \ln f/dz = O(k)$. From (B.8) we conclude that the leading contributions to (2.18) come from the last type of integral, where $f(z)$ is an oscillating function. Such integrals become relatively large because the product of two functions, both oscillating on the scale k^{-1}, contains a term that varies on the macroscopic scale $\gg k^{-1}$, as follows from trigonometric addition theorems. The terms involving the boundary layer function $B^{(1)}_{\sigma,\mathbf{q}_n}$ are seen to be smaller than the leading terms by a factor of the order ε; those containing $B^{(2)}_{\sigma,\mathbf{q}_n}$ are even smaller by a factor $\varepsilon^{3/2}$. This implies, in particular, that the entropy density eigenfunctions, given in (B.1), are negligible for the Brillouin lines.

Inserting now (B.6) and (B.7) into (2.18), thereby neglecting the boundary layer functions as argued above, we obtain with the aid of trigonometric addition theorems

$$\Pi_{\sigma n, -\sigma m}(k_\parallel) = \frac{i\sigma}{(2\pi)^2 d} c(\mathbf{R}, \mathbf{k})\, |\hat{k}_z| \int_{-d/2}^{d/2} dz \frac{dT}{dz} \times \sin\left\{\int_{-d/2}^{z} [Q_{-\sigma z}(z', \mathbf{q}_m) - Q_{\sigma z}(z', \mathbf{q}_n)]\, dz' - i\sigma[b^{(-)}_{-\sigma}(\hat{\mathbf{k}}) + b^{(-)}_{\sigma}(\hat{\mathbf{k}})]\right\} \tag{B.9}$$

From (B.9) is is straightforward to derive Eqs. (4.7)–(4.10), using (3.11), (3.14), (3.15), and (4.5).

APPENDIX C

In this Appendix, we derive Eqs. (4.14)–(4.18). To this purpose, we insert (4.7), (4.11), and (4.12) into (2.16). This yields

$$B_{n\sigma}^{\mathrm{MC}}(\mathbf{k}; R_z) = \frac{\rho(R_z)}{c^2(R_z)} \frac{\pi c(\mathbf{R}, \mathbf{k})}{dc(R_z)} \pi_\sigma(q_{nz}, k_z) \times \frac{1}{\pi} \int_{-d/2}^{d/2} dz \frac{dT}{dz} \sum_m \pi_{-\sigma}(q_{mz}, -k_z)\, G_{m\sigma}(z; \mathbf{R}, \mathbf{k}) \quad \text{(C.1)}$$

where

$$G_{m\sigma}(z; \mathbf{R}, \mathbf{k}) = \frac{1}{\pi} \frac{\exp -i(k_z/|k_z|)[(m-n)\,\pi\chi_1(R_z) - i\sigma\chi_2(R_z)]}{(m-n) + i\sigma(2/\pi)\,\xi(\mathbf{k})} \times \sin[(m-n)\,\pi\chi_1(z) - i\sigma\chi_2(z)] \quad \text{(C.2)}$$

In order to perform the sum in (C.1), we first expand $\pi_{-\sigma}(q_{mz}, -k_z)$ around q_{nz}. Using (4.5), this yields[12]

$$\pi_{-\sigma}(q_{mz}, -k_z) = \sum_{l=0}^{\infty} \frac{1}{l!} \left[(m-n) \frac{\pi c(\mathbf{R}, \mathbf{k})}{dc(R_z)} \right]^l \frac{\partial^l}{\partial q_{nz}^l} \pi_{-\sigma}(q_{nz}, -k_z) \quad \text{(C.3)}$$

Differentiating (C.2) formally with respect to $\chi_1(R_z)$, one obtains on the other hand

$$\frac{\partial^l}{\partial \chi_1(R_z)^l} G_{m\sigma}(z; \mathbf{R}, \mathbf{k}) = \left[-i \frac{k_z}{|k_z|} (m-n)\,\pi \right]^l G_{m\sigma}(z; \mathbf{R}, \mathbf{k}) \quad \text{(C.4)}$$

Combining (C.3) and (C.4) gives

$$\sum_m \pi_{-\sigma}(q_{mz}, -k_z)\, G_{m\sigma}(z; \mathbf{R}, \mathbf{k}) = \sum_{l=0}^{\infty} \frac{1}{l!} \left[i \frac{k_z}{|k_z|} \frac{c(\mathbf{R}, \mathbf{k})}{dc(R_z)} \right]^l \left[\frac{\partial^l}{\partial q_{nz}^l} \pi_{-\sigma}(q_{nz}, -k_z) \right] \times \frac{\partial^l}{\partial \chi_1(R_z)^l} G_\sigma(z; \mathbf{R}, \mathbf{k}) \quad \text{(C.5)}$$

[12] All orders in $(q_{mz} - q_{nz})$ have to be kept, since $\pi_{-\sigma}(q_{mz}, -k_z)$ is sharply peaked, thus depending sensitively on q_{mz}.

where

$$G_\sigma(z; \mathbf{R}, \mathbf{k}) = \sum_m G_{m\sigma}(z; \mathbf{R}, \mathbf{k}) \tag{C.6}$$

Already using here a result from (C.13) below, we can write

$$\frac{\partial^l}{\partial\chi_1(R_z)^l} G_\sigma(z; \mathbf{R}, \mathbf{k}) = \left[-\sigma \frac{k_z}{|k_z|} 2\xi(\mathbf{k})\right]^l G_\sigma(z; \mathbf{G}, \mathbf{k}) \tag{C.7}$$

which allows us to sum the right-hand side in (C.5), yielding

$$\sum_m \pi_{-\sigma}(q_{mz}, -k_z)\, G_{m\sigma}(z; \mathbf{R}, \mathbf{k}) = \pi_{-\sigma}(q'_{nz}, -k_z)\, G_\sigma(z; \mathbf{R}, \mathbf{k}) \tag{C.8}$$

where q'_{nz} is defined in (4.16). Inserting (C.8) into (C.1), we obtain Eqs. (4.14) and (4.15), and it remains to be shown that the sum (C.6) is given by (4.17) and (4.18). To do this, we can restrict ourselves to real admittances $\beta^{(\pm)} \geqslant 0$, since the general case follows by analytic continuation. According to (3.10), (3.12), and (4.10), real admittances imply that the wall absorptions $w^{(\pm)}(\hat{\mathbf{k}})$ have real, nonnegative values. Hence, by (4.9) and (4.13), $\chi_2(z)$ and $\xi(\mathbf{k})$ are real functions. Inserting, after these considerations, (C.2) into (C.6) and replacing the summation index m by $(m-n)$, we can write[13]

$$\begin{aligned} G_\sigma(z; \mathbf{R}, \mathbf{k}) = -\frac{1}{\pi}\left\{\exp\left[-\sigma \frac{k_z}{|k_z|} \chi_2(R_z)\right]\right\} \\ \times\left(\frac{\pi}{2}\frac{\sinh \chi_2(z)}{\xi(\mathbf{k})} - \{\exp[\sigma\chi_2(z)]\}\, G'(\chi_+) \right. \\ \left. + \{\exp[-\sigma\chi_2(z)]\}\, G'(\chi_-)\right) \end{aligned} \tag{C.9}$$

where

$$\chi_\pm = \chi_\pm(z, R_z) = -\frac{k_z}{|k_z|} \pi\chi_1(R_z) \pm \pi\chi_1(z) \tag{C.10}$$

and

$$G'(\chi) = \frac{1}{2i} \sum_{m=1}^{\infty} \frac{e^{i\chi m}}{m + i\sigma(2/\pi)\, \xi(\mathbf{k})} + \text{c.c.} \tag{C.11}$$

[13] We use here that the terms (C.2) are peaked around $m \approx n$, which allows us to extend the sum (C.6) also over negative m values. The error in doing so is of order ε.

The value of the sum in (C.11) is[23]

$$G'(\chi) = \sigma \frac{\pi}{4\xi(\mathbf{k})} + \pi \frac{\exp\{\sigma(2/\pi)\,\xi(\mathbf{k})[\chi - 2\pi l]\}}{1 - \exp[4\sigma\xi(\mathbf{k})]} \tag{C.12}$$

where l is an integer to be chosen such that $0 < \chi - 2\pi l < 2\pi$. If $\chi = 2\pi l$, l integer, the sum (C.11) does not converge. However, this case does not arise here, since $\chi_1(z)$ in (C.10) is by the definition (4.8) a monotonically increasing function of z with values between 0 and 1. Using (C.10) and (C.12) in (C.9) yields after a short calculation

$$\begin{aligned} &G_\sigma(z; \mathbf{R}, \mathbf{k}) \\ &\quad = -\frac{\exp -\sigma(k_z/|k_z|)\{2\xi(\mathbf{k})[\chi_1(R_z) - \Theta(R_z - z)] + \chi_2(R_z)\}}{\sinh 2\xi(\mathbf{k})} \\ &\quad\quad \times \sinh\{2\xi(\mathbf{k})[\chi_1(z) - \Theta(z - R_z)] + \chi_2(z)\} \end{aligned} \tag{C.13}$$

This confirms the result (C.7) we have used above.

From (C.13) it is now easy to find (4.17), (4.18). One merely needs to use (4.19) and the identity

$$2\xi(\mathbf{k})\,\chi_1(z) + \chi_2(z) = 2\xi^{(-)}(z, \mathbf{k}) \tag{C.14}$$

which follows from the definitions (4.8), (4.9), (4.13), and (4.18).

APPENDIX D

In this Appendix, we outline the derivation of the results (4.20) and (4.21) for the line-shape factor. First we insert (4.2), (4.14), and (4.16) into the definition (4.15). This yields a double integral of a form similar to, but slightly more complicated, than Eq. (A.1) in Appendix A. In a manner similar to the calculation there, one substitutes the center-of-mass and relative coordinates as new integration variables. Then the integration over the relative coordinate can be executed straightforwardly. Using also (3.11), (3.15), and (4.10), one is left with

$$\begin{aligned} &\Delta_\sigma^{\mathrm{MC}}(q_{nz} + i\bar{\eta}_\sigma - |k_z|; \mathbf{k}) \\ &\quad = \frac{L_s}{2} \int_{-1/2}^{1/2} d\zeta\, F(\zeta) \exp 2i(q_{nz} + i\bar{\eta}_\sigma - |k_z|)\, L_s \zeta \end{aligned} \tag{D.1}$$

where

$$F(\zeta) = \frac{e^{2\delta\zeta} \sin\{[a\zeta(k_z/|k_z|) - i\delta](1 - 2\,|\zeta|)\}}{a\zeta k_z/|k_z| - i\delta} \tag{D.2}$$

and

$$a = \frac{1}{|\hat{k}_z|} \frac{d \ln c}{dR_z} k L_s^2 \tag{D.3}$$

$$\delta = \sigma \frac{\Gamma_s(R_z)\, k^2}{2c(R_z)\, |\hat{k}_z|} L_s \tag{D.4}$$

Since δ is a small quantity of the order $\sqrt{\varepsilon}$, we expand in (D.2) up to first order in δ:

$$F(\zeta) = \left[1 - i\delta \left(\frac{k_z}{|k_z|} \frac{1}{\zeta} \frac{\partial}{\partial a} + 2i\zeta \right) \right] \frac{\sin a\zeta(1 - 2\,|\zeta|)}{a\zeta} \tag{D.5}$$

Inserting (D.5) into (D.1) yields after some calculation and a partial integration

$$\begin{aligned} &\Delta_\sigma^{\mathrm{MC}}(q_{nz} + i\bar{\eta}_\sigma - |k_z|; \mathbf{k}) \\ &= \frac{1}{\pi} \int_0^{1/2} d\zeta \left\{ f(a, \zeta) \left[1 + i \frac{\delta}{L_s} \frac{\partial}{\partial\, |k_z|} \right] \right. \\ &\quad \left. + \frac{k_z}{|k_z|} \frac{\delta}{2L_s}\, g(a, \zeta) \frac{\partial}{\partial\, |k_z|} \right\} \frac{\sin 2(q_{nz} + i\bar{\eta}_\sigma - |k_z|)\, L_s \zeta}{q_{nz} + i\bar{\eta}_\sigma - |k_z|} \end{aligned} \tag{D.6}$$

where

$$f(a, \zeta) = \frac{\partial}{\partial \zeta} \frac{\sin a\zeta(2\zeta - 1)}{a\zeta} \tag{D.7a}$$

$$g(a, \zeta) = \frac{\partial}{\partial \zeta} \frac{1}{\zeta^2} \frac{\partial}{\partial a} \frac{\sin a\zeta(2\zeta - 1)}{a\zeta} \tag{D.7b}$$

Approximating (D.7) by the leading terms in the Taylor series with respect to a leads with sufficient accuracy in the whole interval $0 \leqslant \zeta \leqslant 1/2$ to

$$f(a, \zeta) \approx 2, \qquad g(a, \zeta) \approx -2a(2\zeta - 1)^2 \tag{D.8}$$

since $a \leqslant O(1)$.[14] Using (D.8) in (D.6), one can evaluate the integral and find the expression (4.20) with $\Delta_1(q)$ and $\Delta_2(q)$ given by (4.4) and (4.21), respectively.

[14] See also the remark in footnote 11.

APPENDIX E

In this Appendix, we summarize the final step in the computation of the Brillouin lines from the nonequilibrium sound modes. This step consists in summing up Eq. (2.13) to obtain the results given in Section 5. Since this part of the calculation is basically the same for the local equilibrium and the mode-coupling part of the Brillouin lines, we will give the details mainly for the local equilibrium part.

Recalling that $L_s k = O(1/\sqrt{\varepsilon}) \gg 1$, we see from (4.4) that $\Delta_1(q_{nz} + i\bar{\eta}_\sigma - |k_z|)$ is concentrated at a peak around $q_{nz} \approx |k_z|$ with width $\sim L_s^{-1}$. Hence, the main contribution to the sum (2.13), with $B_{n\sigma}(\mathbf{k}; R_z)$ given by (4.3), comes from the modes n with q_{nz} lying within that peak around $|k_z|$. For these modes one finds from (3.13), (3.14), and (3.17)

$$s_{\sigma,k_\parallel n} = i\sigma c(R_z)\, Q_\sigma(R_z, \mathbf{q}_n) + \frac{1}{2} \Gamma_s(R_z)\, k^2 \tag{E.1}$$

with

$$Q_\sigma(R_z, \mathbf{q}_n) = [k_\parallel^2 + (q_{nz} + i\bar{\eta}_\sigma)^2]^{1/2} \tag{E.2}$$

In (E.1) and (E.2), we have replaced q_{nz} by $|k_z|$ in the terms that are relatively small, which leads to errors of the order $\sqrt{\varepsilon}$. Inserting now (4.4) and (E.1) into (2.13) yields

$$S_{\mathrm{B}\sigma}^{\mathrm{LE}}(\mathbf{k}, \omega; R_z) = I_{\mathrm{B}}(R_z) \frac{\pi c(\mathbf{R}, \mathbf{k})}{dc(R_z)} \times \sum_n \frac{\Delta_1(q_{nz} + i\bar{\eta}_\sigma - |k_z|)}{i[\sigma c(R_z)\, Q_\sigma(R_z, \mathbf{q}_n) - \omega] + \frac{1}{2}\Gamma_s(R_z)\, k^2} + \text{c.c.} \tag{E.3}$$

where we have also used the identity (5.2).

In (E.3), the distance between adjacent levels q_{nz} and $q_{n+1,z}$ is of the order $1/d$ [cf. Eq. (E.4) below], i.e., smaller by a factor $\sqrt{\varepsilon}$ than the width ($\sim 1/L_s$) of the peak. Thus, the q_{nz} can be considered to lie quasidensely, and, since the terms in (E.3) are smoothly varying functions of q_{nz}, we can approximate the sum by an integral. Since the density of states is, from (4.5),

$$q_{n+1,z} - q_{nz} = \frac{\pi c(\mathbf{R}, \mathbf{k})}{dc(R_z)} \tag{E.4}$$

we obtain from (E.3), using also (E.2),

$$S_{\mathrm{B}\sigma}^{\mathrm{LE}}(\mathbf{k}, \omega; R_z) = I_{\mathrm{B}}(R_z)\, \mathrm{Re} \int_0^\infty dq_z' \frac{2\Delta_1(q_z' + i\bar{\eta}_\sigma - |k_z|)}{i\{\sigma c(R_z)[k_\parallel^2 + (q_z' + i\bar{\eta}_\sigma)^2]^{1/2} - \omega\} + \frac{1}{2}\Gamma_s(R_z)\, k^2} \tag{E.5}$$

In (E.5) we can replace the lower bound of the integral by $-\infty$, since the added tail $-\infty < q'_z \leqslant 0$ lies far outside the main peak of Δ_1. Substituting then $Q'_z = q'_z + i\bar{\eta}_\sigma$ leads to an integral in the complex plane over a straight line $\mathscr{C}$ shifted parallel to the real axis by the amount Re $\bar{\eta}_\sigma$. Using Re $\beta^{(\pm)} \geqslant 0$, one can easily show from (3.10)–(3.12), (3.15), and (E.5) that the integrand has no pole between $\mathscr{C}$ and the real axis. Hence one can shift the path of integration back onto the real axis and obtain

$$S^{\mathrm{LE}}_{\mathrm{B}\sigma}(\mathbf{k}, \omega; R_z) = I_{\mathrm{B}}(R_z)\,\mathrm{Re} \int_{-\infty}^{\infty} dQ'_z \frac{2\Delta_1(Q'_z - |k_z|)}{i[\sigma c(R_z)(k_\parallel^2 + Q'^2_z)^{1/2} - \omega] + \frac{1}{2}\Gamma_s(R_z)\,k^2} \tag{E.6}$$

Finally, we use once more that $Lk = O(1/\sqrt{\varepsilon}) \gg 1$ to approximate the sharply peaked function $\Delta_1(q)$ by a delta function:

$$\Delta_1(q) = \delta(q) \tag{E.7}$$

In fact, the right-hand side in (E.7) is the first term in a multipole expansion of $\Delta_1(q)$, due to the normalization chosen in the definition (4.4). When (E.7) is inserted into (E.6) one finds immediately that the integral is equal to $Y_{\mathrm{B}\sigma}(\mathbf{k}, \omega; R_z)$, thus verifying the expressions (5.1)–(5.3).

For the mode-coupling part the calculation is similar, since also the line-shape factor $\Delta^{\mathrm{MC}}_\sigma(q_{nz} + i\bar{\eta}_\sigma - |k_z|; \mathbf{k})$, defined in (4.20), is sharply peaked around $q_{nz} \approx |k_z|$. The normalization in (4.21) has been chosen such that

$$\Delta_2(q) = \delta(q) \tag{E.8}$$

inour approximation. Thus, we find from (2.13), (4.14), (4.20), (E.1), (E.7), and (E.8) in a similar manner

$$S^{\mathrm{MC}}_{\mathrm{B}\sigma}(\mathbf{k}, \omega; R_z) = \mathrm{Re}\left\{ I^{\mathrm{MC}}_{\mathrm{B}\sigma}(\mathbf{k}; R_z) \left[1 + \sigma \frac{1}{c(R_z)\,|\hat{k}_z|}\, \Omega(\mathbf{k}; R_z) \frac{\partial}{\partial\,|k_z|} \right] \times Y_{\mathrm{B}\sigma}(\mathbf{k}, \omega; R_z) \right\} \tag{E.9}$$

where we have also used (5.3), (5.5), and (5.7). Our expression (5.6) is obtained from (E.9) by making use of the equation

$$\frac{\partial}{\partial\,|k_z|} Y_{\mathrm{B}}(\mathbf{k}, \omega; R_z) = -\sigma c(R_z)\,|\hat{k}_z| \frac{\partial}{\partial\omega} Y_{\mathrm{B}}(\mathbf{k}, \omega; R_z) \tag{E.10}$$

which follows from (5.3), neglecting corrections of order ε.

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