INFLUENCE OF EVAPORATION AND CONDENSATION UPON SOUND REFLECTION

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Abstract-The reflectance r of a liquid surface for sound incident from the saturated vapour depends upon the evaporation coefficient σ , which offers a possibility of measuring this coefficient. For a sufficiently dense vapour and for sound frequencies well below the collision frequency in the vapour, hydrodynamic equations combined with appropriately modified boundary conditions can be applied to analyse the effect. A strong dependence of r upon σ is predicted for frequencies not too small in comparison with the reciprocal of a relaxation time, which for water at 20° C equals 1.8×10^{-6} s. The dependence is slightly uncertain and indirect, given through the intermediary of a phenomenological jump coefficient L_{nn} .

NOMENCLATURE

- A, B, dimensionless amplitudes of sound waves at $x=0$;
- c, speed of sound (low-frequency limit);
- c_p , specific heat at constant pressure
- (per unit mass) ;
- D , $= \lambda/\rho c_p$, thermal diffusivity;
- j, evaporating mass flux;
 $k, \infty/c$, wave number of
- k, $\approx \omega/c$, wave number of sound wave;
 K , $\approx (T/p) d p/dT$, coefficient from
- $= (T/p) dp/dT$, coefficient from Clausius-Clapeyron equation;

 L_{pp} , etc., phenomenological jump coefficients in bulk evaporation;

- M_{\odot} molecular mass ;
- equilibrium vapour pressure at p, temperature *T;*
- heat of vaporization (per unit mass) ; q, reflectance;
- r. R,
- molar gas constant ; specific entropy; \overline{s} ,
- time ; \bar{t} .
- T. temperature (undisturbed value);
- bulk velocity ; μ ,
- $= (8RT/\pi M)^{1/2}$, average molecular \tilde{v} , speed ;
- $\mathbf{x},$ distance from surface $(x > 0$ within vapour) ;
- Z, specific acoustic admittance of surface.

Greek symbols

- β , volume expansion coefficient;
 γ , = c_n/c_v, ratio of specific heats
- $= c_n/c_v$, ratio of specific heats;
- $\varepsilon_p, \varepsilon_T$, relative pressure and temperature jumps;

$$
\Delta p, \Delta T, \Delta \rho, \Delta s
$$
, excess pressure, temperature, density, specific entropy;

- θ , angle of incidence;
- dimensionless amplitude of heat wave Θ , at $x=0$;
- $\approx (\omega/2D)^{1/2}$, wave number of heat wave; κ
- heat conductivity ; λ,
- $=$ $(\eta_{\text{bulk}} + \frac{4}{3}\eta)/\rho$, longitudinal kinematic $\mathbf{v},$ viscosity;
- density (equilibrium value at *T);* ρ ,
- evaporation coefficient ; σ .
- τ, characteristic relaxation time of liquid-vapour system ;
- $\approx D_1/c_1^2$, mean free time between τ_c
- collisions of vapour molecules ; $+ 1$ A T ¹

$$
\zeta, \qquad = \Delta p/p + \frac{1}{2}\Delta T/T, \text{ jump parameter};
$$

 $2\pi \times$ sound frequency. ω ,

Subscripts

- 1, vapour;
2, liquid.
- liquid.

1. THE EVAPORATION COEFFICIENT

IN EQUILIBRIUM between a liquid and its saturated vapour, as long as the latter can be considered as an ideal gas, the mass flux of molecules crossing the boundary each way is given by the familiar expression $\frac{1}{4}\sigma\bar{v}\rho_1$, where $\bar{v} = (8RT/\pi M)^{1/2}$ = $(8/\pi\gamma_1)^{1/2}c_1$ is the average molecular speed. The evaporation coefficient σ specifies the fraction of molecules incident from the vapour that merge with the liquid, while the fraction $1-\sigma$ refers to molecules scattered back into the vapour $\lceil 1, 2 \rceil$. If the incident part of the molecular distribution remains a Maxwellian at rest, yet with the corresponding pressure and temperature modified to $p + \Delta p_1$ and $T + \Delta T_1$, the

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the Hertz-Knudsen formula, the towards acoustic frequencies has been made [9].

$$
j = -\frac{1}{4}\sigma\bar{v}\rho_1\xi,
$$

\n
$$
\xi = \frac{\Delta p_1}{p} - \frac{1}{2}\frac{\Delta T_1}{T}.
$$
\n(1)

A more refined description must take into account the scattering probability w of the incident molecules, which depends upon their velocity. According to detailed balance, the equilibrium average of w equals $1 - \sigma$ [2]. As practically nothing is known about the velocity distribution after scattering, two extreme models may be considered: (a) perfect accommodation, i.e. the scattered molecules have a Maxwellian distribution in equilibrium with the surface, which implies that w is velocity independent; and (b), specular reflection. Reality is likely to be closer to the first extreme.

One must keep in mind that formula (1) only applies to a free-molecular regime under the stated specific conditions. As was shown by several authors and as will be explained in Section 3, under steadystate hydrodynamic conditions the coefficient is roughly twice as large-a fact sometimes overlooked in the evaluation of experiments. This coefficient is essential in predictions of growth rates of bubbles in a superheated liquid and of droplets in a supersaturated vapour.

It is disappointing to learn that, because of experimental difficulties, evaporation coefficients and the corresponding hydrodynamic coefficients are poorly known, especially at higher vapour pressures. For water at ordinary temperature, the values compiled by Cammenga et al. [3] range from 0.002 to 1. Careful measurements recently carried out by Springer [4] gave 0.2 for a surface continuously renewed by overflow. A five times smaller value resulted if the surface was stagnant and therefore presumably more contaminated. It is widely believed that the true σ is close to unity, and some observations with liquids at lower vapour pressures seem to support this opinion [5,6]. Further investigation appears to be needed.

Contamination should be easier to avoid in a fast experiment, which implies a non-steady state. It is then imperative that changes of the surface temperature can be assessed unambiguously. Measurements of sound-wave reflection offer such a possibility. Experiments of this kind have first been made by Maurer [7], on the basis of an unpublished proposal by Volmer and a theory by Becker and Döring. However, thermal effects have been neglected in the evaluation. A more complete analysis has been carried out by Meinhold-Heerlein for the coupling of sound to second sound at the surface of superfluid helium [8]. It should also be mentioned that a related method is being used since some time for investigating adsorption/desorption rates. Though only very slow oscillations (a few cycles per minute or less) produced by moving pistons have been

evaporating net mass flux is approximately given by applied so far, a suggestion to extend the range

While low-frequency sound impinging from a noncondensing gas upon the surface of a liquid or solid is almost totally reflected, some dissipation has been observed with ultrasound. The effect has been explained by Herzfeld as being due to heat waves generated at the interface $[10, 11]$. Enhanced dissipation must be expected for sound incident from a vapour upon its own condensed phase, because of flow through the interface and exchange of latent heat. The subsequent analysis is intended to derive, under somewhat idealized conditions, the dependence of the reflectance upon frequency and the evaporation coefficient.

2. SOUND AND HEAT WAVES

In order to stay within the hydrodynamic regime we are going to choose a sound frequency several orders below the molecular collision frequency (τ_c^{-1}) in the vapour, which for water at 20°C is about 3 \times 10⁸ Hz. A sufficient description is then given by the changes Δp , $\Delta \rho$, ΔT , Δs of pressure, density, temperature and specific entropy, and by the bulk velocity u in both media. The normal to the surface, pointing into the vapour, will be taken as the x axis, with the surface at $x = 0$. Only plane waves of normal incidence will be considered, so that Δp $= \Delta p(x, t)$ etc.

The acoustic disturbance consists of an incident wave, reflected wave, and transmitted wave. In addition, we have two heat waves generated at the interface where latent heat is periodically liberated and absorbed. If amplitudes are small, as will be assumed, the two kinds of waves are linearly superposed. Their modes of propagation are determined by linearized hydrodynamic equations (heat conduction, Navier-Stokes, continuity) for a longitudinal disturbance $[11, 12]$,

$$
\lambda \frac{\partial^2 \Delta T}{\partial x^2} = \rho T \frac{\partial \Delta s}{\partial t},\qquad(2)
$$

$$
\frac{\partial u}{\partial t} = -\frac{1}{\rho} \frac{\partial \Delta p}{\partial x} + v \frac{\partial^2 u}{\partial x^2},
$$
 (3)

$$
\frac{\partial u}{\partial x} = -\frac{1}{\rho} \frac{\partial \Delta \rho}{\partial t},\tag{4}
$$

supplemented by the linearized equations of state,

$$
\Delta s = (c_p/T)\Delta T - (\beta/\rho)\Delta p, \qquad (5)
$$

$$
\Delta \rho = (\gamma/c^2) \Delta p - \beta \rho \Delta T.
$$
 (6)

Substituting an exponential dependence upon x and *t, we* derive both branches of the dispersion relation and the amplitude ratios. Sound waves propagating in the directions $\pm x$ are represented by

$$
\left[\frac{\Delta p}{p}, \frac{u}{c}, \frac{\Delta T}{T}\right] = \left\{1, \pm \frac{p}{\rho c^2} \left[1 - \frac{i\omega}{2c^2} (v - D')\right],
$$
\n
$$
\frac{p}{\rho c^2} \frac{\gamma - 1}{\beta T} \left(1 - \frac{i\omega}{c^2} D\right)\right\}
$$
\n
$$
\times A \exp\left(\pm ikx - i\omega t\right),
$$
\n(7)

$$
k = \frac{\omega}{c} \left[1 + \frac{i\omega}{2c^2} \left(v + D' \right) \right],\tag{8}
$$

where $D' = (\gamma - 1)D$; and the heat waves by

$$
\begin{aligned}\n\left[\frac{\Delta p}{p}, \frac{u}{c}, \frac{\Delta T}{T}\right] &= \left[-i\omega \frac{\beta T \rho}{p} \left(v - D\right), \mp (1 - i)\beta T \left(\frac{\omega D}{2c^2}\right)^{1/2}, 1\right] \\
&\times \Theta \exp\left[\mp (1 - i)\kappa x - i\omega t\right], \quad (9)\n\end{aligned}
$$

$$
\kappa = \left(\frac{\omega}{2D}\right)^{1/2} \left[1 + \frac{i\omega}{2c^2}(\gamma - 1)(\nu - D)\right].\tag{10}
$$

Only first-order corrections have been incorporated to account for sound absorption and for a related effect in heat waves. For monatomic vapours, smallness of these corrections is guaranteed by the previous assumption since $\omega v_1/c_1^2 \sim \omega D_1/c_1^2 \approx \omega \tau_c$ \ll 1. On the other hand, in polyatomic gases such as water vapour, absorption can be many times stronger because of internal (mainly vibrational) relaxation $\lceil 11-13 \rceil$. Phenomenologically, this is accounted for by a relatively large bulk viscosity. If $\omega v_1/c_1^2$ is not small enough, a hydrodynamic description requires the introduction of further quantities, e.g. of different temperatures for the various degrees of freedom. To avoid such difficulties we will require that always $\omega v_1/c_1^2 \ll 1$. Therefore, when dealing with boundary conditions in the next sections, the respective terms as well as those with the factor $\omega D_1/c_1^2$ [but not the one with $(\omega D_1/c_1^2)^{1/2}$] will be dropped from equations (7-10).

3. HYDRODYNAMIC BOUNDARY CONDITIONS

The assumption made with equation (1) implies that the vapour density is small ($\rho_1 \ll \rho_2$) and allows us to neglect the sound wave entering the liquid, as it carries only a small fraction of the incident energy ($\sim 0.002\%$ for water at 20°C). It will be consistent to consider the whole liquid as well as the interface as being at rest, in spite of the mass exchange and the resulting bulk flow of the vapour. Hence the evaporating mass flux is given by

$$
j(t) = \rho_1 u_1(0, t). \tag{11}
$$

Four boundary conditions connect the solutions for the vapour to those for the liquid. The first, given by the continuity of pressure,

$$
\Delta p_1 = \Delta p_2 \quad \text{at} \quad x = 0,
$$

will not be needed, as we are not interested in Δp_2 . Next, we observe that the heat of evaporation must be supplied by heat currents from both media, thus

$$
\lambda_1 \frac{\partial \Delta T_1}{\partial x} - \lambda_2 \frac{\partial \Delta T_2}{\partial x} = \rho_1 u_1 q \text{ at } x = 0. \quad (12)
$$

It would be wrong to take continuity of temperature and the Hertz-Knudsen formula (1) for the remaining two boundary conditions. Because of the resulting bulk motion of the vapour, and because of transition effects, the molecular distribution is modified. Readjustment of the distribution takes place within the Knudsen layer, whose thickness is of the order of one mean free path for monatomic vapours, or correspondingly larger for polyatomic vapours (because of internal relaxation). The layer may be regarded as a weak stationary shock front. Only after transversing the shock does the vapour attain local thermodynamic equilibrium with a bulk velocity u_1 and a unique temperature T_1 . If this temperature is extrapolated down to the surface, a discontinuity in form of a temperature jump must be expected. Such extrapoiated values already appear in equation (12) and will be considered henceforth.

The relative deviation of vapour pressure from saturation at the temperature $T+\Delta T_2$ is given by ε_p $= [\Delta p_1 - (Kp/T)\Delta T_2]/p$, where *K* is the dimensionless coefficient from the Clausius-Clapeyron equation,

$$
K = \frac{T}{p} \frac{dp}{dT} = \frac{Mq}{RT}.
$$

For small departures from equilibrium the pressure deviation as well as the temperature jump are linearly related to the bulk velocity and to the temperature gradient in the vapour near $x = 0$ [8, 14, 151. Let us state these relations as

$$
\varepsilon_{p} \equiv \frac{\Delta p_{1}}{p} - K \frac{\Delta T_{2}}{T} = \frac{\gamma_{1}}{c_{1}} \left[-L_{p\mu} u_{1} + L_{p\tau} \frac{\lambda_{1}}{p} \frac{\partial \Delta T_{1}}{\partial x} \right],
$$

\n
$$
x = 0, \quad (13)
$$

\n
$$
\varepsilon_{T} \equiv \frac{\Delta T_{1} - \Delta T_{2}}{T} = \frac{\gamma_{1}}{c_{1}} \left[-L_{p\tau} u_{1} + L_{\tau\tau} \frac{\lambda_{1}}{p} \frac{\partial \Delta T_{1}}{\partial x} \right],
$$

\n
$$
x = 0, \quad (14)
$$

where L_{pp} etc. are the appropriate phenomenological jump coefficients. Their normalization differs here from Lang's [15] by a trivial factor :

$$
L_{pp} = (1/2\gamma_1)^{1/2} L_p^{(M)}
$$
 etc.

The assumption that $\omega \tau_c \ll 1$ and also $\omega v_1/c_1^2 \ll 1$ is again essential. Only then is the Knudsen layer able to follow the changes in both media quasistatically, so that the coefficients L_{pp} etc. are frequency independent.

In order to make a comparison with equation (1) we take $\Delta T = 0$ and $\partial \Delta T / \partial x = 0$. Since $\xi = e$ $-\frac{1}{2}\varepsilon_T$, the mass flux is given by

$$
\rho_1 u_1 = -\left(\frac{\pi}{8\gamma_1}\right)^{1/2} \frac{1}{L_{pp} - \frac{1}{2}L_{pT}} \bar{v}\rho_1 \xi. \tag{15}
$$

This can be considered as the hydrodynamic analog of the Hertz-Knudsen formula (1). Except at very low vapour pressures, steady-state observations directly yield the coefficient in equation (15) and not $\frac{1}{2}$.

All three phenomenological coefficients depend in a subtle way upon the evaporation coefficient, as well as upon differential scattering probabilities both for intermolecular collisions and coIlisions with the surface. A rough approximation is obtained by Maxwell's method $\lceil 1 \rceil$, which consists in extrapolating the incident distribution from the hydrodynamic region down to the surface. If the molecules scattered by the surface are perfectly accommodated the results are

$$
L_{pp} = \left(\frac{\pi}{2\gamma_1}\right)^{1/2} \left[\frac{2-\sigma}{\sigma} + \frac{1}{2}\frac{\gamma_1 - 1}{\gamma_1 + 1}\right],\tag{16}
$$

$$
L_{pT} = \left(\frac{\pi}{2\gamma_1}\right)^{1/2} \frac{\gamma_1 - 1}{\gamma_1 + 1} = \frac{1}{2} L_{TT}.
$$
 (17)

Remarkably, L_{pT} and L_{TT} do not depend upon σ in this approximation, which is expected to involve errors of the order of 10% . The corresponding coefficient in equation (15) is seen to differ from that in the Hertz-Knudsen formula by the factor $2/(2-\sigma)$ $[16, 17]$.

Values believed to be accurate within a few percent follow through a variational method previously used for monoatomic vapours $\lceil 15, 18 \rceil$. For the cases of perfect accommodation (a), and of specular reflection with velocity~independent scattering probability fb), these approximations are

$$
L_{pp} = \left(\frac{\pi}{2\gamma_1}\right)^{1/2} \left\{ \left[\frac{2}{\pi} + \frac{1+3\gamma_1}{4(1+\gamma_1)} \right] + \frac{2}{\sigma} (1-\sigma) \right\},\qquad(18a)
$$

$$
L_{pp} = \left(\frac{\pi}{2\gamma_1}\right)^{1/2} \left\{ \left[\frac{2}{\pi} + \frac{1+3\gamma_1}{4(1+\gamma_1)} \right] + \left[\frac{2}{\pi} + \frac{1+3\gamma_1}{4(1+\gamma_1)} \frac{4-\sigma}{\sigma} \right] (1-\sigma) \right\}. \quad (18b)
$$

4. DERIVATION OF THE REFLECTANCE

With neglect of terms of relative order $\omega v_1/c_1^2$ and $\omega D_1/c_1^2$, the sound field in the vapour and the two heat waves are described by the real parts of the following expressions,

$$
\frac{\Delta p_1}{p} = (A e^{-ik_1x} + B e^{ik_1x}) e^{-i\omega t}, \qquad (19)
$$

$$
\frac{u_1}{c_1} = \left\{ -\frac{1}{\gamma_1} \left(A e^{-ik_1 x} - B e^{ik_1 x} \right) - (1 - i) \left(\frac{\omega D_1}{2c_1^2} \right)^{1/2} \Theta_1 \exp[-(1 - i)\kappa_1 x] \right\}
$$

× $e^{-i\omega t}$, (20)

$$
\frac{\Delta T_1}{T} = \left\{ \Theta_1 \exp\left[-(1-i)\kappa_1 x \right] + \frac{\gamma_1 - 1}{\gamma_1} \left(A e^{-ik_1 x} + B e^{ik_1 x} \right) \right\} e^{-i\omega t}, \quad (21)
$$

$$
\frac{\Delta T_2}{T} = \Theta_2 \exp[(1-i)\kappa_2 x] e^{-i\omega t}.
$$
 (22)

Substitution into the boundary conditions (12) - (14) yields a homogeneous system of equations determining the ratios of the complex amplitudes A, B, Θ_1 , $\Theta_2,$

$$
(1-i)(\lambda_1\kappa_1\Theta_1 + \lambda_2\kappa_2\Theta_2)T
$$

=
$$
\frac{\rho_1qc_1}{\gamma_1} \left[(A-B) + (1-i)\gamma_1 \left(\frac{\omega D_1}{2c_1^2}\right)^{1/2} \Theta_1 \right], \quad (23)
$$

$$
(A+B)-K\Theta_2
$$

= $L_{pp}(A-B)+(1-i)\left(\gamma_1 L_{pp} - \frac{\gamma_1^2}{\gamma_1 - 1} L_{pT}\right)$

$$
\times \left(\frac{\omega D_1}{2c_1^2}\right)^{1/2} \Theta_1, (24)
$$

$$
\frac{\gamma_1 - 1}{\gamma_1} (A+B)+\Theta_1 - \Theta_2
$$

= $L_{pT}(A-B)+(1-i)\left(\gamma_1 L_{pT} - \frac{\gamma_1^2}{\gamma_1 - 1} L_{TT}\right)$

$$
\times \left(\frac{\omega D_1}{2c_1^2}\right)^{1/2} \Theta_1, (25)
$$

For any specific set of data a numerical solution of these equations can be produced at once, and the reflectance $r = |B/A|^2$ derived therefrom.

We see by inspection that B, Θ_1 and Θ_2 have at most the same order of magnitude as *A.* Hence, for not too large L_{pp} (not too small σ) a simple albeit rough approximation follows by neglecting in equations (23) and (24) also the terms involving Θ_1 , where the coefficients are of the order $(\omega \tau_c)^{1/2}$. Equation (25) thereby becomes superfluous, as we are only interested in the ratio B/A . This means that we end up ignoring the heat wave in the gas. Essentiafly the heat wave in the Iiquid and the sound waves in the vapour determine the reflectance.

To descrihe the final result in the familiar form

$$
r = |B/A|^2 = |(1 - Z)/(1 + Z)|^2, \tag{26}
$$

we defme a specifx acoustic admittance of the surface by $[19]$

$$
Z = \frac{i}{k} \left(\frac{1}{\Delta p} \frac{\partial \Delta p}{\partial x} \right)_{\text{sound.}} = \frac{A - B}{A + B}.
$$

After eliminating Θ , from the simplified equations (23) and (24) , we obtain the approximation

$$
Z^{-1} = L_{pp} + \frac{1}{(1-i)(\omega \tau)^{1/2}},
$$
 (27)

where

$$
\tau = \omega^{-1} \left(\frac{\gamma_1 \lambda_2 \kappa_2 T}{\rho_1 q c_1 K} \right)^2 = \frac{\gamma_1 \lambda_2 \rho_2 c_{p2} RT^3}{2 \rho_1^2 M q^4}.
$$
 (28)

The parameter τ introduced here represents a relaxation time characteristic for the liquid-vapour system. Very roughly, τ is inversely proportional to the square of the saturated vapour density; for water at 20 $^{\circ}$ C it amounts to 1.8×10^{-6} s. A useful observation inferred from expression (28) is that $\tau/\tau_c \gg 1$ as long as $\rho_1 \ll \rho_2$.

At lower vapour pressure, τ may well exceed the reciprocal frequencies of applied sound. In the limit of $\omega \tau \ge 1$, expression (27) simplifies to $Z^{-1} = L_{nn}$, so that

$$
r = \left[(L_{pp} - 1)/(L_{pp} + 1) \right]^2. \tag{29}
$$

Apart from a different approximation for $L_{\text{p}p}$, this is the formula used by Maurer $[7]$. However, the applied frequency was too low and the vapour pressure too high to justify the simplification.

By way of illustration, we substitute L_{pp} from equations (18a) and (18b), and the value $y_1 = 1.33$ for water. When σ increases from 0 to 1, L_{pp}^{-1} is seen to increase from 0 to 0.785 which makes r to decrease according to equation (29) from 1 to 0.0145. At lower frequencies the range of r slowly shrinks, as shown by Fig. 1, and in the limit as $\omega \tau \rightarrow 0$, the reflectance approaches unity, regardless of the value of L_{nn} . This was to be expected, as the whole system is able to adjust itself to very slow changes in a quasistatic manner.

FIG. 1. Dependence of the reflectance upon frequency and the jump coefficient L_{pp} , according to equations (26) and (27). The lower and upper scales for σ are according to equations (18a) and (18b), respectively, both for $\gamma_1 = 1.33$.

At moderately low frequencies the effect is still noticeable and allows to distinguish the extremes σ = 0 and σ = 1. However, for σ not sufficiently small the sensitivity $\partial r/\partial L_{pp}$ falls off like $(\omega t)^{3/2}$ so that acoustic measurements soon become worthless. The corresponding flat portions of the curves on Fig. 1 hardly depend upon L_{pp} and can be approximated by $r \approx 1 - 4(\omega \tau)^{1/2}$.

For water at 100°C and with a frequency of 3.5 kHz Maurer obtained a reflectance below 0.4, which is much less than the value predicted by equations (26), (27). Damping due to water adsorbed on the wall of the experimental tube could perhaps explain the discrepancy.

The analysis is easily generalized to oblique incidence. Except for grazing angles the result is $\lceil 19 \rceil$

$$
r = |(\cos \theta - Z)/(\cos \theta + Z)|^2. \tag{30}
$$

At a certain angle of incidence, r is seen to reach a minimum, which in the high-frequency limit is $r = 0$ at $\theta = \arccos(1/L_{pp})$.

For very small σ , when L_{pp} becomes large, the derivation of equation (27) becomes inaccurate. Since in this case $\Theta_1 \ge A - B$ and $\Theta_1 \ge \Theta_2$, one must retain some of the terms involving Θ_1 . If bulky expressions are to be avoided, the best is to extract a rough approximation for Θ , from the previously omitted equation (25),

$$
\Theta_1 = -\left[\left(\gamma_1 - 1 \right) / \gamma_1 \right] (A + B),
$$

and to substitute this into equations (23) and (24). The resulting admittance is

$$
Z = Z_0 \Biggl\{ 1 - K \frac{\gamma_1 - 1}{\gamma_1} \frac{\lambda_1 \kappa_1}{\lambda_2 \kappa_2} + (1 - i) \times \left(\frac{\omega D_1}{2c_1^2} \right)^{1/2} \biggl[(\gamma_1 - 1) Z_0^{-1} - \gamma_1 L_{pT} \biggr] \Biggr\}, \quad (31)
$$

where Z_0 stands for the value from equation (27). In the limit as $\sigma \to 0$, $L_{pp} \to \infty$, $Z_0 \to 0$, the result agrees with the one calculated by Herzfeld [10] simply from the boundary condition $u_1 = 0$ at $x = 0$:

$$
Z = (1 - i)(\gamma_1 - 1)(\omega D_1/2c_1^2)^{1/2}
$$

$$
r \approx 1 - 4Re(Z).
$$
 (32)

5. COMMENTS

As was shown, heat and mass exchange at the vapour-liquid interface can strongly influence the reflection of sound. If this is to be exploited for measurements of the phenomenological jump coefficient L_{pp} , frequencies are needed such that $\omega\tau$ is not too small, yet at the same time $\omega \tau_c \ll 1$ and also $\omega v_1/c_1^2 \ll 1$. Hence the ratio $v_1/c_1^2 \tau$ must be small, say $\lesssim 0.1$, which for polyatomic vapours imposes a severe restriction upon permissible vapour pressures. It appears doubtful whether with water vapour, which exhibits strong relaxation [12], the method could be applied much beyond 50°C where $p = 0.12$ bar.

The values of L_{pp} and L_{pT} are all we need to predict evaporation and condensation rates under near-equilibrium hydrodynamic conditions. However, the quantity representing a genuine surface property is σ , which has to be derived from the measured L_{pp} . An error of the order $0.1(1-\sigma)$, as inferred by comparing equations (18a) and (18b), is inevitable in this derivation as long as the distribution of scattered molecules remains unknown.

Hydrodynamic boundary conditions cannot be used at very low pressures, such as found with nonvolatile liquids. There the mean free path in the vapour may become longer than applicable wavelengths of sound, or even comparable to the dimensions of the apparatus. For such a regime the analysis would be tedious, and could hardly produce unambiguous results. Only in the extreme case of a collisionless gas is the situation simple again [20]. One expects the reflectance in this case to be proportional to $1-\sigma$, though an uncertainty of several tens of per cent is involved because of the unknown surface-scattering law. Perhaps the equipment developed by Greenspan and others [21-23] for acoustic studies of rarefied gases could be used under such circumstances.

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INFLUENCE DE L'EVAPORATION ET DE LA CONDENSATION SUR LA REFLEXION **DU SON**

Résumé—Le pouvoir réfléchissant r d'une surface liquide vis-à-vis du son incident dans la vapeur saturée dépend du coefficient d'évaporation σ , ce qui offre une méthode pour mesurer ce coefficient. A une densité de vapeur suffisante et pour des fréquences sonores faibles par rapport à la fréquence de collision dans la vapeur, on peut analyser l'effet par les équations hydrodynamiques associées à des conditions aux limites modifiées de façon appropriée. Pour des fréquences pas trop faibles en comparaison avec l'inverse d'un temps de relaxation (égal à 1,8·10⁻⁶s pour l'eau à 20°C), on prévoit une forte dependance de *r* vis-a-vis de σ . La dépendance donnée au moyen d'un coefficient phénoménologique de saut L_{pp} est un peu incertaine et indirecte.

EINFLUSS VON VERDAMPFUNG UND KONDENSATION AUF SCHALLREFLEXION

Zusammenfassung-Das Reflexionsvermögen r einer Flüssigkeitsoberfläche gegenüber Schall, der aus dem gesättigten Dampf auftrifft, hängt vom Verdampfungskoeffizienten σ ab und bietet somit eine Moglichkeit **zur** Messung dieses Koeffizienten. Fur geniigend hohe Dampfdichten und fur Schallfrequenzen, die klein gegen die Stoßfrequenz im Dampf sind, können zur Analyse des Effekts hydrodynamische Gleichungen mit entsprechend modifizierten Randbedingungen verwendet werden. Eine starke Abhängigkeit der Größe r von σ wird für Frequenzen, die nicht zu klein sind verglichen mit dem Kehrwert einer Relaxationszeit, vorausgesagt. Hierbei erhält man für Wasser von 20°C eine Relaxationszeit von 1.8 $\times 10^{-6}$ s. Die Abhängigkeit des r von σ ist mit einer Ungewißheit behaftet und indirekt, über einen phänomenologischen Sprungkoeffizienten L_{pp} gegeben.

ВЛИЯНИЕ ИСПАРЕНИЯ И КОНДЕНСАЦИИ НА ОТРАЖЕНИЕ ЗВУКА

Аннотация - Отражательная способность г поверхности раздела жидкость - насыщенный пар при падении на неё звуковой волны зависит от коэффициента испарения жидкости σ , что позволяет измерять этот коэффициент. Для достаточно плотного пара и частоты звука, HaMHOLO Меньшей частоты соударения частиц в паре, при анализе этого явления можн использовать гидродинамические уравнения с соответствующими граничными условиями. Предсказана сильная зависимость r от o при частотах, величина которых не слишком мала по сравнению с величиной, обратной времени релаксации, составляющей 1,8 х 10⁻⁶ сек для воды при 20 С. Эта зависимость определяется с помощью феноменологического коэффи-**UHeHTa L,,,.**