Experimental Detection of Forces Produced by the Flow of Heat

C. Albanese,[1] P. Dell'Aversana,[1] and F. S. Gaeta[1,2,*]
[1]*MARS Center, Via Comunale Tavernola, 80144 Napoli, Italy*
[2]*IIGB of CNR, Via Marconi 10, 80125 Napoli, Italy*

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We detected thermal radiation forces produced by heat flow through solid slabs suspended in nonisothermal liquids. Lateral effects are avoided, allowing a unidimensional approach. The results confirm expectations. Numerically simulated gravity-dependent perturbations are smaller than the measured forces. More precise measurements will be attempted on a European Space Agency suborbital flight in 1998. [S0031-9007(97)04544-4]

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The existence in liquids of thermal excitations consisting of high frequency wave packets was postulated by Debye in 1912 [1]. Naturally occurring hyperfrequency waves were experimentally detected fifty years later, by their interactions with light [2–6], and the low frequency part of the spectrum of thermal waves in liquids could be explored. Propagation velocities, up to about 10 GHz, are only slightly higher than those of ordinary sound.

More recently, collective, longitudinal excitations in a much higher frequency range, where wavelengths become comparable with intermolecular distances, were observed in liquid D_2O at room temperature by coherent inelastic neutron scattering [7]. Their propagation velocity is $3310 \pm 250\ \mathrm{m\,s^{-1}}$. By a different technique, based on inelastic x-ray scattering, Sette *et al.* [8,9] were able to study thermal waves in liquid H_2O in an energy-momentum region wider than the one accessible to neutron scattering in D_2O. Naturally occurring ultrahigh frequency elastic waves exhibited a solidlike measured velocity of $3200 \pm 100\ \mathrm{m\,s^{-1}}$. From these studies a fundamental analogy emerges in the dynamical responses of liquid H_2O and D_2O with those of the respective ices.

A fundamental difference between the properties of wavelike thermal excitations in solids and liquids should, however, be kept in mind, due to the ubiquitous presence of disorder in dense fluids over mesoscopic distances. This has the consequence of distorting the harmonic potential within which the molecules oscillate. In consequence of the skewness of the potential wells, the oscillations of the particles are anharmonic. Furthermore, there will be a multiplicity of nearby modes of slightly different energy. Interactions among such anharmonic modes are well known in solids [10].

We propose that liquids may be viewed as dual systems constituted by a "gas" of thermal excitations and a population of particles, interacting with a mutual exchange of energy and momentum.

I. Thermal radiation forces: a quantitative evaluation.—In the course of the interactions, when the system is isotropic and homogeneous, energy and momentum may be gained or lost with equal probability by the gas of excitations and the local groups of oscillating particles. Only when a symmetry-breaking factor such as a temperature gradient is applied does the energy and momentum exchange among excitations and particles become unbalanced. In these conditions, the drift of wave packets along the temperature gradient causes, in the average, an excess transfer of energy and momentum to the particles.

Neglecting the tensorial nature of thermal conductivity, we shall consider a unidimensional case. The momentum flux $\mathbf{J}_p$ coupled to a flux of thermal energy $\mathbf{J}_q$ will be $\mathbf{J}_p = \mathbf{J}_q/u_g$, u_g being the group velocity of propagation of the wave packets. Inelastic scattering processes transfer momentum to the medium at an *a priori* unknown rate, producing a volume force, i.e., a pressure gradient in the medium:

$$\left(\frac{\partial P}{\partial z}\right)^{\mathrm{th}}\mathbf{r} = \frac{\partial}{\partial z}(H\mathbf{J}_p) = -\frac{\partial}{\partial z}\left(H\,\frac{K}{u_g}\,\frac{dT}{dz}\right)\mathbf{r}\,, \qquad (1)$$

$\mathbf{r}$ being the unit vector in the direction of the temperature gradient, K is the thermal conductivity, and H is a numerical quantity ranging between zero and unity, which is the reflection coefficient of mechanical waves [11–14]. If the rate of momentum transfer to the medium is exceedingly small ($H \approx 0$), there will be no observable pressure gradient.

Let us consider a system in which the heat flows across a surface of discontinuity between two adjacent media—say, two immiscible liquids or a solid and a liquid. The heat flux is continuous through the limiting surface, but there is a discontinuity in the momentum flux, due to the abrupt change of the propagation velocity of thermal waves—an extreme case of anharmonicity. From Eq. (1), a thermal radiation pressure is produced at the interface, given by

$$\begin{aligned}\mathbf{P}^{\mathrm{th}}_{1,2} &= \Delta_{1,2}(H\mathbf{J}_p) = H_{1,2}\left[\left(\frac{\mathbf{J}_q}{u_g}\right)_2 - \left(\frac{\mathbf{J}_q}{u_g}\right)_1\right]\mathbf{r} \\ &= H_{1,2}\left\{\left(K\,\frac{dT}{dz}\right)\left[\left(\frac{1}{u_g}\right)_1 - \left(\frac{1}{u_g}\right)_2\right]\right\}\mathbf{r}\,, \qquad (2)\end{aligned}$$

where subscripts 1 and 2 refer to the heat-emitting and heat-receiving surface. It should be observed that the sign of the thermal force per unit of surface depends on whether $(1/u_g)_2 \gtrless (1/u_g)_1$.

An expression analogous to Eq. (2) was derived by an extension of Rayleigh's concept of acoustic radiation pressure to nonisothermal systems [11]. Subsequently, it was deduced from an extension of a Boltzmann theorem and from considerations of rational mechanics [12]. These approaches have led to a physically coherent interpretation of thermomechanical and mechanothermal effects in liquids and to new insight on the issue of microscopic time reversibility [13,14].

II. Experimental.—To validate the theory, we measured the forces produced by heat flowing through a solid disk immersed in a nonisothermal liquid. According to the theory, forces will be produced at the liquid-solid upper interface, as well as at the lower interface, over all of the disk surface crossed by the heat flow. (Here $\mathbf{P}^{\mathrm{th}}_{1,2}$ should be understood as $\mathbf{P}^{\mathrm{th}}_{l,s}$, the media being liquid and solid.) The above approximation holds until the advancing temperature front reaches the lateral surface of the disk.

Experimentation with various couples of solids and liquids may, thus, yield a check of the existence of thermal radiation forces (TRFs) and their *modus operandi*. Information may also be obtained on the propagation velocities of wave packets through Eq. (2), which, however, requires accurate, perturbation-free measurements, impossible on ground.

A dedicated experimental apparatus, sketched in Fig. 1, has been designed to measure TRFs. The measuring device has been realized by integrating an analytical balance having a force sensitivity of about 10^{-1} dyn, with a thermostated container filled with a liquid in which the slab is suspended. A heater, formed by a Ni-Cr wire wrapped around a copper cylinder, dipping in the liquid, is fixed above the slab. Bath and heater temperatures are controlled independently; the heater temperature can rise up to 90 °C. The slab is connected with the balance by means of a couple of very thin tungsten wires (diam $= 50\ \mu$m). Care has been used to reduce undesired effects; in our apparatus, the horizontal position of the slab and its parallelism with the heater surface are optically ensured within ±20 min of arc, sufficient to achieve the desired reproducibility in the tests. The distance between the slab and the heater has been fixed with an accuracy of 0.1 mm.

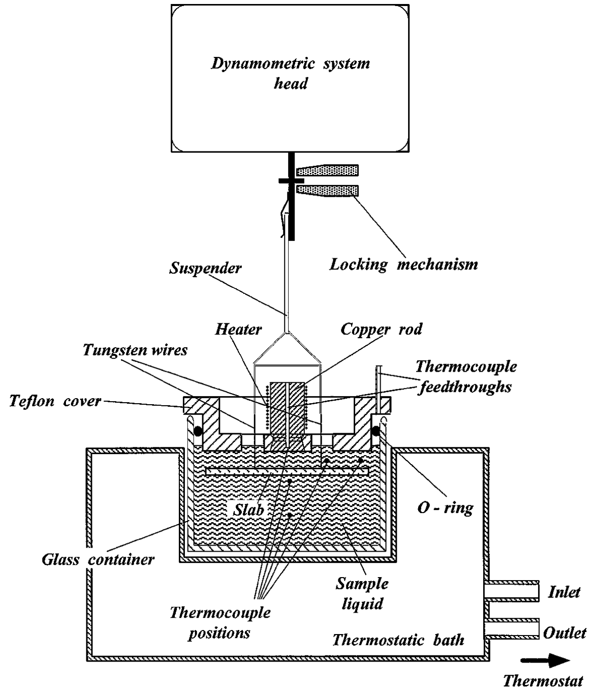
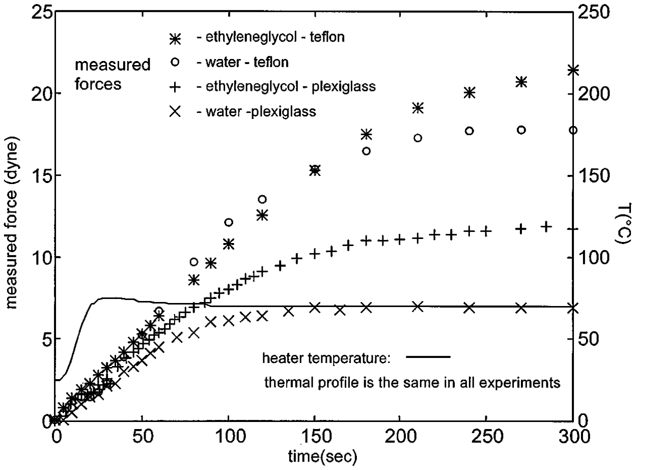
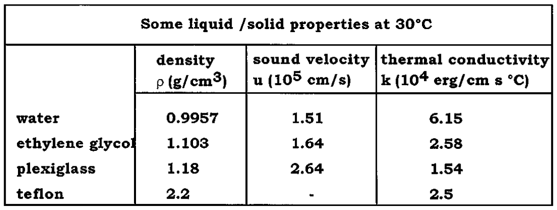


FIG. 1. Schematic representation of the apparatus used for the measurement of the TRFs on ground.

The liquids used in the experiments are water, ethylene glycol, glycerol, and Fluorinert®, while slabs have been made with Teflon® and Plexiglas®. The geometry of the apparatus ensures that border effects are avoided at least for a few minutes. Temperature readings are performed by thermocouples distributed in key points inside the experimental cell with particular attention to the space near the solid disk.

As the heater is switched on, the temperature rises following an imposed ramp until it reaches, after a short overshoot, the imposed temperature. Simultaneously, the force measurements are started; a timer is used as a common time base for the heater temperature profile and the signals coming from the thermocouples inserted in the liquid. In Fig. 2, the heater temperature profile and the force signals have been plotted for solid disks, 4 cm in





Some liquid /solid properties at 30°C			
	density ρ (g/cm³)	sound velocity u (10⁵ cm/s)	thermal conductivity k (10⁴ erg/cm s °C)
water	0.9957	1.51	6.15
ethylene glycol	1.103	1.64	2.58
plexiglass	1.18	2.64	1.54
teflon	2.2	-	2.5

FIG. 2. Force and temperature profiles in two experiments with disks 4 cm in diameter of Plexiglas® (0.2 cm thick) and Teflon® (0.3 cm thick) in water and in ethylene glycol. The temperature profile and the initial temperature (+20 °C) are always the same. Some properties of the materials are listed; sound velocity refers to ultrasonic frequencies.

diameter, immersed in various liquids. (Since the force is always negative, i.e., opposite to $\mathbf{J}_q$, we refer to its absolute value.)

After an initial steep rise of both the temperature and force profiles, the forces continue to increase for a while, after the heater temperature has reached the steady plateau level. The force profile, indeed, follows the building up of the temperature gradient across the disk, through an area which widens while the heat diffuses to an increasingly large region around the heat source. A plateau value of the force is reached, followed, under normal gravity, by a slow decrement, due to the decrease of the intensity of local temperature gradient with time, to the decreasing buoyancy in the progressively warmer liquid and, finally, to the onset of lateral effects, when the advancing temperature front reaches the border of the disk. Experiments with various solid-liquid couples evidenced a delay of the force signal increasing with the distance from the heater, and dependent on the heat diffusivity of the materials. The decrease of the forces measured after switching off the heating also fits well the approach discussed so far. One example of a registration inclusive of the phase after the switch off of the heat source is given in Fig. 3.

The reproducibility of results is fair, typical variations in distinct runs executed under the same experimental conditions being congruous with the fluctuations in the course of a single experiment, amounting to less than 5% of the measured forces.

On ground, the various perturbing factors are difficult to assess, owing to the overlap of all of the gravity-induced disturbances. An evaluation of the most important perturbing hydrodynamical effects is now presented. Applying heat from above, convection is established in the liquid gap between the heater and the solid slab, and between the hot zone beneath the heater and the surrounding cold zone near the border of the slab. Convective motions have been observed by introducing a dye, methylene blue, laterally, near the border of the heater; they generate a force pulling the disk up, i.e., in the same direction of the TRF. From a sequence of images, the mean convective velocity of 0.07 cm/sec in the radial direction was deduced.

Another disturbance due to the change of buoyancy comes from the different thermal expansions of liquid and solid. From the instantaneous temperature distribution, this force contribution, Δf_{buoy}, may be approximately calculated at any instant. Finally, capillary forces, which pull the tungsten wires down, decrease as the upper surface of the liquid warms up; a differential force, pointing up, thus appears at the beginning of a measurement in the first part of the heating phase and disappears during the plateau. The hydrostatic pressure variation connected with thermal expansion of the heated liquid gives equal contributions on the upper and lower surfaces of the slab, so that it has no net effect on the measured force. These effects act more or less independently; each contributes a force normal to the surface of the disk, some pushing it up, others pulling down as shown in Fig. 4. The resultant, thus, is given by the algebraic sum of the respective components.

A numerical simulation has been done to evaluate the system evolution from the fluid-dynamic point of view. The geometry used in the simulation is axisymmetric, and reproduces quite well the experimental cell geometry. Using the simulation results, the entity of the convective effects near the heater and the buoyancy variations, due to the heating of the liquid surrounding the solid slab, have been calculated. We found that the contribution of TRF to the measured force is preeminent, and that the measured total forces represent the intensity of the respective TRF with a 10% overall margin of imprecision.

III. Discussion and conclusions.—An extensive discussion of the theoretical implications of our findings would be out of place here, since this paper is a report

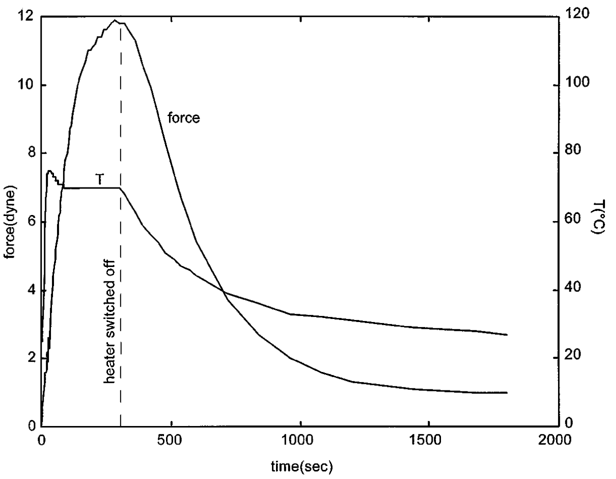


FIG. 3. Temperature and force profiles in an experiment with a Plexiglas® disk 4 cm in diameter and 0.2 cm thick in ethylene glycol. The post-heater switch-off events are shown.

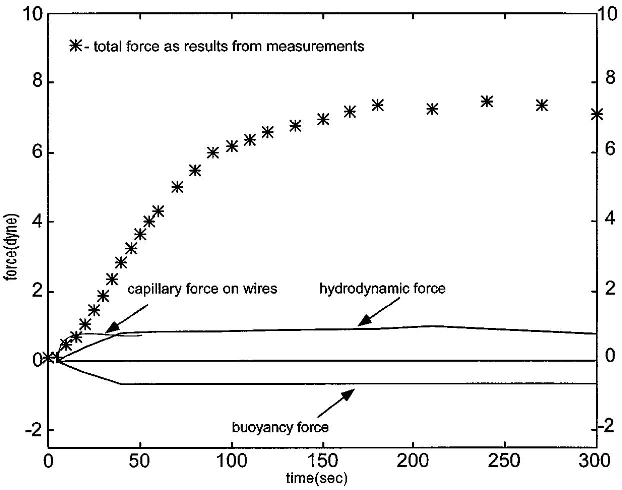


FIG. 4. Hydrodynamic force, buoyancy force, and capillary effect on the suspender wires, calculated with the aid of numerical simulations, together with the total measured force on a Plexiglas® disk 4 cm in diameter and 0.2 cm thick, in water.

on preliminary experimental measurements. However, we would like to make a concise comment of a general nature.

The existence of well defined—though strongly damped—sound waves, of frequencies up to very high values, i.e., to wavelengths comparable with interatomic distances, was evidenced by de Schepper *et al.* also in liquid argon [15]. Compiling these results with those from neutron scattering and x-ray scattering in associated hydrogen-bonded liquids such as D_2O and H_2O mentioned above [7–9], it seems possible to conclude that thermal excitations of frequencies extending up to the 1000 GHz range exist in all types of liquids. Their propagation speed at very high frequencies ("fast sound") might be substantially higher than group velocity in the ultrasonic range. Solidlike velocities are not surprising, in view of the similarity of the values of intermolecular forces at short range in the two states of aggregation, which suggest analogy of dynamical responses of liquids and solids to periodic solicitations having mesoscopic wavelengths.

From the experiments performed until now, it is already possible to assess the existence and order of magnitude of the TRF, notwithstanding the presence of various perturbations of gravitational origin. The analysis of our results only allows calculation of approximate values for the TRF of each solid-liquid couple of materials. These values may be compared with the ones derived from Eq. (2) (last expression) inserting the constitutive properties of the materials and the experimental conditions. The propagation velocities u_g which can be used in the calculation are those measured at sonic or ultrasonic frequencies, three or four orders of magnitude lower than those of thermal wave packets. Inserting these data in Eq. (2), together with the other pertinent constitutive properties of the materials employed and the values of dT/dz, we can calculate the expected values of the TRFs. These turn out to be of the same order of magnitude as the experimental data.

In conclusion, the detection of TRFs with all of the tested solid-liquid couples of materials is confirmed. The coupling of thermal energy with a flux of momentum is thus proved. The imprecision of measurements, unavoidable on ground, is particularly unfortunate since the results are not good enough to allow the derivation of a precise value for the propagation velocity of hyperfrequency elastic waves in liquids. On the other hand, with our method, there are no limitations concerning the nature of the liquids employed. Thus, a very extensive investigation of the dynamical behavior of the liquid state at mesoscopic lengths appears feasible, if gravity-dependent perturbations may be eliminated. The method *per se* doesn't meet any of the serious limitations encountered by coherent inelastic neutron [7,15] or inelastic x-ray scattering [8,9].

Further experimentation will be carried out in conditions of weightlessness on a European Space Agency suborbital Maser flight scheduled for the spring of 1998.

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*Person to whom all correspondence should be addressed.

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