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# Thermal conductivity associated to the bead-bead contact decorated by a liquid bridge

## An experimental study based on the response of a chain subjected to thermal cycles

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**Abstract.** We report an experimental study of the heat transport along a chain of macroscopic beads: at one end, one bead is periodically heated and we record the resulting temperature variations of another bead as a function of position and time. The experimental results show that the chain behaves like a high-order low-pass filter. The measurement of the associated cut-off frequency makes possible to determine accurately the resistance of the bead-bead contact: we make use of the experimental setup for studying the effects of liquid bridges decorating the bead-bead contacts, which provides important clues for understanding the thermal properties of partially wet granular matter.

**PACS.** 66.70.+f Nonelectronic thermal conduction and heat-pulse propagation in solids; thermal waves – 81.05.Rm Porous materials; granular materials – 91.60.Ki Physical properties of rocks and minerals; thermal properties

## 1 Introduction

The transport of electricity, sound or heat in a dense assembly of solid particles addresses interesting and puzzling questions. Even if granular materials are widely used in industry, one is still far from understanding entirely how the transport coefficients relate with the physics of the contacts between the particles and the geometry of the contact network (*texture*). For instance, for over a century [1,2], electrical transport in metallic powders has generated interest. These systems exhibit fascinating properties, such as highly non-linear and hysteretic electrical conductivities as well as extreme sensitivity to electromagnetic waves [3]. The propagation of soundwaves in granular materials also exhibits very interesting features; nonlinearities such as a slight dependence of the transmission amplitude on the frequency of the source at low vibration amplitude and an extreme sensitivity to the position of each grain [4]. The propagation of soundwaves is so sensitive to the texture that it is likely to be altered by the vibration itself or by small changes in the temperature. However, the propagation of non-linear waves in a one-dimensional chain of beads under static stress has been experimentally proven to follow the prediction of Hertz theory as long as plastic deformations of the grains at the contacts are not involved [5].

In the same way, the transport of heat in granular materials addresses puzzling questions which are relevant to be answered as thermal properties of granular matter are of great practical importance. Indeed, the most common

materials used to insure thermal as well as acoustic insulation in buildings consist in assemblies of solid particles or fibers. For instance, glasswools are efficient thermal insulators that also present the advantage to be low-cost, impurities and fire resistant as they are produced mainly from recycled glass and sand [6]. Moreover, because of their ductility, they can absorb any unevenness of the substrate. Another important application worth to be mentioned here is the use of granular materials as buffers in nuclear-waste deposits [7]. Experimental and theoretical studies of the heat transport in granular materials have already been reported [2,9–11]. They pointed out the crucial role played by the physics of the contacts between the grains. Accordingly, humidity has been experimentally proven to increase significantly the thermal conductivity as water tends to decorate the contact points [12].

Whereas this latter study considered the thermal properties of the bulk granular material, involving thus the texture of the material, there does not exist, to our knowledge, any experimental study dedicated to the thermal properties of the decorated contacts alone. In the present article, we report an experimental study of the thermal conductivity of a one-dimensional chain of beads in the same spirit as preceding studies of electric conductivity [3] and sound propagation [5] that made it possible to analyze the physics of the contacts in absence of texture effects. The response of the system to temperature cycles shows that a periodic medium can be an efficient thermal-insulator in the sense that, for instance, the chain is very

efficient in filtering temperature changes as it acts as a high-order low-pass filter. We determine the corresponding cut-off frequency in order to obtain reliable measurements of the thermal resistance associated to the bead-bead contact. We make use of the experimental setup for studying the effects of liquid bridges decorating the bead-bead contacts.

The content of the manuscript is organized as follows: in the section 2, we introduce the principle of the experiment and describe the experimental setup and procedure. In the section 3, we report our experimental results obtained in the cases of dry or wet contacts and compare to theoretical predictions. Finally, we conclude in the section 4.

## 2 Experimental setup and procedure

The experiment consists in heating periodically one end of a linear chain of beads and in recording the resulting temperature of the  $n^{\text{th}}$  bead as a function of time  $t$ . In the next section 2.1, we describe the experimental setup whereas the section 2.2 is dedicated to the procedure used to analyze the thermal response of the system to the heat injection (Fig. 1).

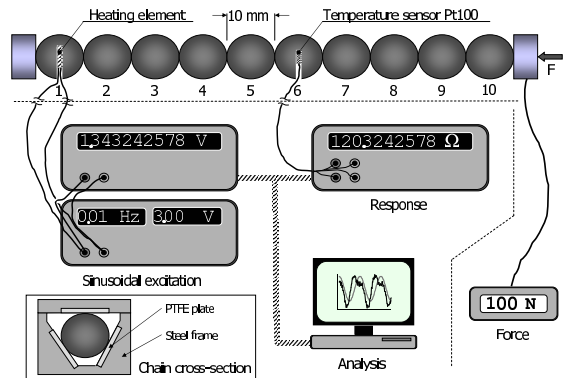
### 2.1 Experimental setup

The chain consists of 10 centimetric ball-bearing steel-beads (AISI 304, diameter  $d = 1$  cm, from Marteau & Lemarié) aligned with the help of three PTFE plates [see the chain cross-section in the inset (Fig. 1)] that insure small thermal contact with the remaining part of the experimental setup which consists of a stainless steel frame. The contact between the beads is insured by pushing (static axial force  $F$ ) the chain at one end against a fixed steel cylinder located at the other end. A force sensor (Entran, ELA-B2E-10KN) is used to measure  $F$  to within 10 N ( $F < 10$  kN).

In order to inject heat at one end of the chain, the first bead is equipped with a small heating wire (Constantan, diameter 100  $\mu\text{m}$ , typical resistance  $r \simeq 1 \Omega$ ) placed at its center. We impose the periodic voltage  $U(t) = U_0(1 + \sin 2\pi\nu t)$  from a home-made linear power-amplifier driven by a function generator (Stanford Research Systems, DS345). A first multimeter (Keithley, 2001) is used to measure the voltage difference  $U(t)$  across the heat source (Fig. 2). In our experimental conditions, the frequency  $\nu$  ranges from 1/3600 to 1/30 Hz whereas the maximum voltage  $2U_0$  is of about 3 V. The power  $P(t)$  injected in the first bead thus writes:

$$P(t) = P_0 \left[ \frac{3}{4} + \sin 2\pi\nu t + \frac{1}{4} \sin \left( 4\pi\nu t + \frac{3\pi}{2} \right) \right] \quad (1)$$

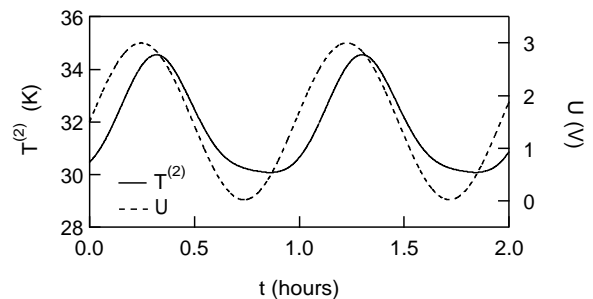
where  $P_0 = 2U_0^2/r$  (we neglect here the variation of  $r$  with the temperature). We point out that the bead is continuously heated with the mean power  $\frac{3}{4}P_0$  and that the variation with time of the heating power  $P(t)$  contains two



**Fig. 1. Sketch of the experimental principle.**

The chain consists in 10 centimetric steel-beads align with the help of three PTFE plates (inset). In order to insure the contact between the beads, a static force  $F$  is applied at one end (bead 10). The first bead (bead 1) is heated by means of a small resistive wire located at its center: the heating voltage is measured by means of a first multimeter. In order to obtain the local temperature of the chain, the bead  $n$  ( $n = 6$  in the sketched situation) is equipped with a Pt-sensor which resistance is measured with the help of a second multimeter. In addition, the temperature of the surrounding air, between beads 7 and 8, is measured by means of a second Pt-sensor (not sketched in this figure).

harmonics. We shall make use of this latter property for studying simultaneously the response of the system at the three frequencies 0,  $\nu$  and  $2\nu$ .



**Fig. 2. Experimental  $T^{(2)}$  and  $U$  vs. time  $t$ .**

We report, for  $\nu = 1/3600$  Hz, the temperature  $T^{(2)}$  of bead 2 and the voltage  $U$  as functions of time  $t$  during 2 hours in the permanent regime (the total duration of the experiment is 10 hours, corresponding to 10 periods of the excitation). The mean temperature is about 2.5 K above the temperature  $T_a$  of the surrounding air ( $\delta T_0^{(2)} \simeq 2.5$  K). The amplitude of the temperature variations at  $\nu$  is about 2.21 K whereas it is about 0.46 K at  $2\nu$  (dry contacts,  $F = 100$  N).

The bead  $n$  (the index  $n$  refers to the position of the bead in the chain) is equipped with a temperature sensor (Pt100, from Heraeus) located at its center. A second multimeter (Keithley, 196), used in 4-wires configuration, measures the resistance  $R_s$  of the sensor so that the local temperature of bead  $n$ ,  $T^{(n)}(t)$ , is known to within about

$10^{-3}$  K. As we do not regulate the overall temperature of the experimental setup, we measure, in addition, the outside air temperature,  $T_a$ , by means of a second temperature sensor (Pt100) located in air and connected to a third multimeter (Keithley, 196).

The function generator and the three multimeters are connected to a computer through a IEEE interface. For given experimental conditions (position  $n$  of the sensing bead, applied force  $F$ , dry or wet contacts), a simple C-routine automatizes the experiment: for a chosen set of heating powers  $P_0$  and frequencies  $\nu$ , the voltage  $U(t)$  and the temperature  $T^{(n)}(t)$  are measured (sampling frequency 1 Hz) and saved to the hard-drive (Fig. 2). Analysis of the data is performed afterwards (Igor Pro 4, Wave-Metrics, Inc.).

## 2.2 Transfer function

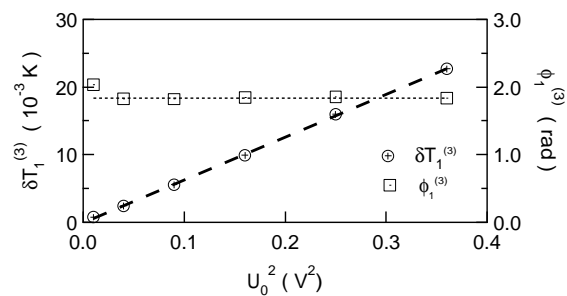
From equation (1), if the system has a linear response, we expect the temperature  $T^{(n)}(t)$  to exhibit the same spectral components as  $P(t)$ , this is to say, harmonics at frequencies 0,  $\nu$  and  $2\nu$ . From the raw data (Fig. 2), we extract five relevant quantities; the amplitudes,  $\delta T_1^{(n)}$  and  $\delta T_2^{(n)}$ , the phases,  $\phi_1^{(n)}$  and  $\phi_2^{(n)}$ , of the two harmonics at the frequencies  $\nu$  and  $2\nu$  and the mean temperature difference  $\delta T_0^{(n)}$  between the bead  $n$  and the outside atmosphere (0 frequency). To do so, we interpolate the experimental temperature-difference  $\Delta T^{(n)}(t) \equiv T^{(n)}(t) - T_a$  with

$$\Delta T^{(n)}(t) = \delta T_0^{(n)} + \delta T_1^{(n)} \sin(2\pi\nu t + \phi_1^{(n)}) + \delta T_2^{(n)} \sin(4\pi\nu t + \phi_2^{(n)}). \quad (2)$$

The origin of time  $t$  in equation (2) is accurately obtained by interpolating  $U(t)$  with a sine-function and by choosing the origin such as  $U(0) = U_0$  and  $\frac{dU}{dt}(0) > 0$ .

We first check that the response of the system is linear: the amplitude of each of the harmonics,  $\delta T_{1,(2)}^{(n)}$ , scales like  $U_0^2$  (thus, is proportional to the heating power) whereas the corresponding phase,  $\phi_{1,(2)}^{(n)}$ , does not depend on  $U_0$  (Fig. 3).

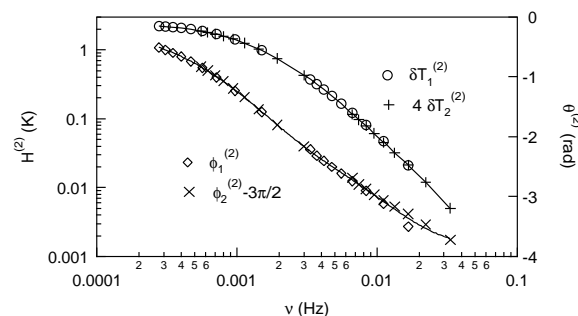
Thus, from the quantities  $\delta T_0^{(n)}$ ,  $\delta T_1^{(n)}$ ,  $\delta T_2^{(n)}$ ,  $\phi_1^{(n)}$  and  $\phi_2^{(n)}$ , we can determine experimentally the power-to-(temperature of the bead  $n$ ) transfer-function,  $\underline{H}^{(n)}(\nu) \equiv H^{(n)}e^{j\theta^{(n)}}$ , as follows: a first set of data points is obtained from the first harmonics at the frequency  $\nu$ . From the first harmonics, we get  $H^{(n)} = \delta T_1^{(n)}$  and  $\theta^{(n)} = \phi_1^{(n)}$ . In equation (1), we notice that the power injected in the first harmonics at  $\nu$  is 4 times larger than that injected in the second harmonics at  $2\nu$ , and that there is phase-shift  $\frac{3\pi}{2}$  between them. Thus, from the same experiment, the second harmonics provides another set of data points as  $H^{(n)} = 4\delta T_2^{(n)}$  and  $\theta^{(n)} = \phi_2^{(n)} - \frac{3\pi}{2}$  at the frequency  $2\nu$ . Note that, by construction,  $H^{(n)}$  has the dimension of temperature and is proportional to  $P_0$ . Strictly speaking, we should divide  $H^{(n)}$  by  $P_0$  to get the power-to-(temperature of the bead  $n$ ) transfer-function, but, as we



**Fig. 3. Amplitude  $\delta T_1^{(3)}$  and phase  $\phi_1^{(3)}$  vs.  $U_0^2$ .**

As an example, we report the amplitude,  $\delta T_1^{(3)}$ , and phase,  $\phi_1^{(3)}$ , measured for bead 3 as functions of the square heating-voltage  $U_0^2$ . The amplitude of the harmonics  $\delta T_{1,(2)}^{(n)}$  is proportional to  $U_0^2$  and the phase  $\phi_{1,(2)}^{(n)}$  does not depend on  $U_0$ , which proves that the response of the system is linear.

will not change  $P_0$  in what follows, we will report  $H^{(n)}$  as the temperature defined above. In figure 4, we report the typical experimental result for  $\underline{H}^{(2)}(\nu)$ . We observe that the data evaluated from the first and second harmonics superpose down to temperature variations of amplitude about  $10^{-3}$  K.



**Fig. 4. Experimental  $H^{(2)}$  and  $\theta^{(2)}$  vs. frequency  $\nu$ .**

As an example, we report the amplitude,  $H^{(2)}$ , and the phase,  $\theta^{(2)}$ , as functions of the frequency  $\nu$ . The data evaluated from the first or the second harmonics respectively are marked with different symbols. We observe that the responses estimated from the first and second harmonics respectively superpose down to temperature variations of amplitude about  $10^{-3}$  K (the lines are only guides for the eye).

We shall explain in the section devoted to the experimental results (section 3) how the response of the chain can be extracted from these data, provided we are able to model the response of the experimental setup.

## 2.3 Experimental procedure

The experimental procedure is as follows: for chosen experimental conditions (applied force  $F$ , dry or wet contacts), in order to obtain the response of the system, we first place the bead containing the temperature sensor at a chosen position  $n$ . For a series of excitation frequencies

$\nu$ , we record the temperature variation  $T^{(n)}$  as a function of time  $t$  in the permanent regime. The response at a different position  $n$  is obtained after opening the experimental setup and changing the position of the sensing bead. The experimental procedure makes possible to determine  $\underline{H}^{(n)}(\nu)$  for  $n \in [2, 7]$  in the frequency range  $1/\nu \in [30, 3600]$  s. Measurements at frequencies smaller than  $1/3600$  Hz are, in principle, possible but note that we record the temperature variations for 10 periods, which then would lead to an experimental time larger than 10 hours. For frequencies larger than  $1/30$  Hz, the temperature variations become small (amplitude typically less than  $10^{-3}$  K) and the response of the system is determined with poor accuracy. We always make sure that the measurements are reliable by checking that the phase of the measured harmonics is locked (*i.e.* that the phase is not arbitrary with respect to that of the heating voltage) and exclude the data points that do not satisfy this condition.

### 3 Experimental results

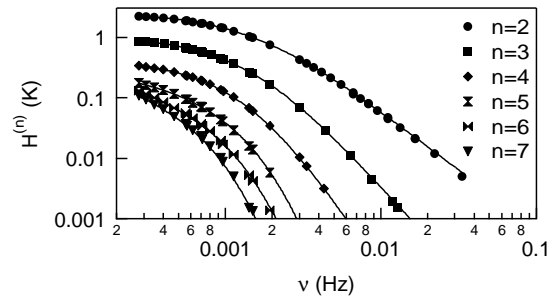
In the following, we make use of the experimental setup for studying the thermal properties of the chain in different experimental conditions. In the next section 3.1, we consider the case of dry contacts in air: the analysis of the whole set of experimental data makes possible the modeling of the thermal response of the whole experimental setup. Then, section 3.2 is dedicated to the study of wet contacts obtained by addition of liquid droplets in the bead-bead contact-regions.

#### 3.1 Dry contacts

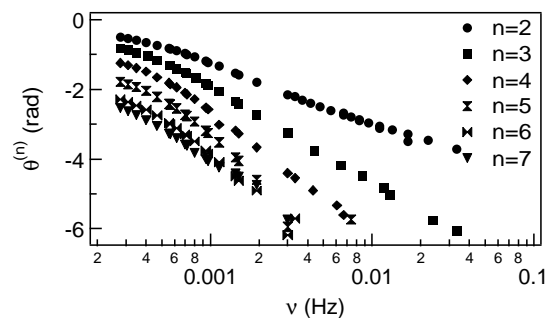
##### 3.1.1 First experimental observations

In figures 5 and 6, we present the whole set of experimental data obtained for the chain of steel beads with dry contacts and  $F = 100$  N (the static applied force is small and only insures that the beads are at contact). At first sight, the chain behaves like a low-pass filter which order depends on the position  $n$  of the bead within the chain (the larger  $n$  is, the larger is the order of the filter). Nevertheless, we cannot immediately extract the characteristic frequency of the filter without exploring in details the response of the experimental setup.

For instance, consider only the response of the system at zero frequency. Due to thermal losses, one would expect the mean temperature  $\delta T_0^{(n)}$  (accordingly  $H^{(n)}(0)$ ) to vanish (*i.e.*  $T^{(n)}(t) \rightarrow T_a$ ) for  $n \rightarrow \infty$ . However, even if we observe that  $H^{(n)}(0)$  decreases almost exponentially when  $n$  is increased, the mean temperature-difference  $H^{(n)}(0)$  surprisingly does not vanish for large  $n$  (Fig. 7). The exponential decay of  $H^{(n)}(0)$  to a finite temperature-difference is the most obvious experimental manifestation of the non-trivial response of the experimental setup. It turns out that, the response at zero frequency is not enough for us

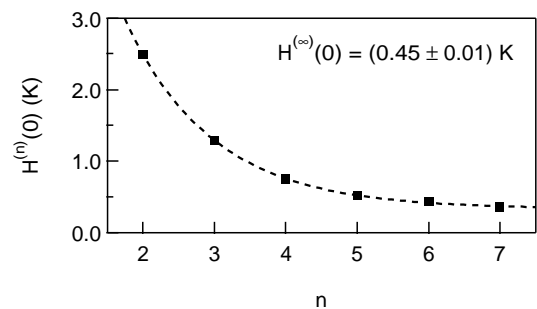


**Fig. 5. Amplitude  $H^{(n)}$  vs. frequency  $\nu$ .** The amplitude  $H^{(n)}$  decreases faster with increasing frequency  $\nu$  for larger  $n$  ( $F = 100$  N, dry contacts, the lines are only guides for the eye).



**Fig. 6. Phase  $\theta^{(n)}$  vs. frequency  $\nu$ .** The absolute phase-shift  $\theta^{(n)}$  increases faster with increasing frequency  $\nu$  for larger  $n$  ( $F = 100$  N, dry contacts).

to measure the thermal losses to the outside atmosphere: the whole set of experimental data is necessary for obtaining a correct model of the experimental situation, which we discuss in the next section.



**Fig. 7. Experimental  $H^{(n)}(0)$  vs. bead position  $n$ .** We observe that the temperature difference at zero frequency between the bead  $n$  and the atmosphere decreases almost exponentially when  $n$  increases but does not vanish far away from the heat source (large  $n$ ).

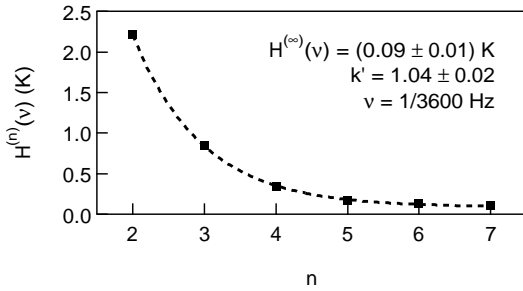
##### 3.1.2 Dynamical response

From the experimental data presented in figure 5, we can extract the dependance of the amplitude  $H^{(n)}$  on the po-

sition  $n$  at a given frequency  $\nu$  (Fig. 8). We observe again that, even at non-zero frequency, the amplitude  $H^{(n)}$  decreases exponentially when  $n$  increases but does not vanish for large values of  $n$ . We write:

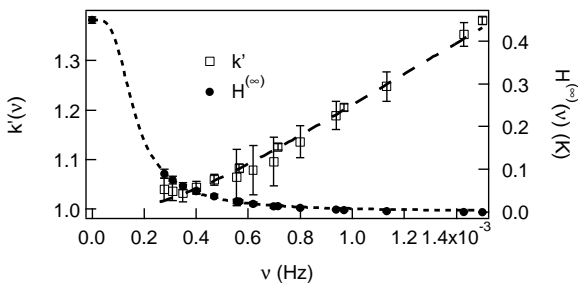
$$H^{(n)}(\nu) = H^{(0)}(\nu) \exp[-k'(\nu)n] + H^{(\infty)}(\nu) \quad (3)$$

and report in figure 9 the experimental values of  $H^{(\infty)}(\nu)$  [Eq. (4)] and  $k'(\nu) = \Re(k)$  [Eq. (5)] which can be accounted by the modeling of the experimental situation as follows (Fig. 10): We consider the beads as heat capacitors (heat capacity  $C$ ) connected one to its two neighbors through thermal resistors (resistance  $R$ ), and we assume that the thermal losses to the frame can be associated to the thermal resistance,  $\beta$  along the chain ( $n \geq 2$ ) and  $\gamma$  at bead 1. Detailed description of the model is proposed in the appendix.



**Fig. 8. Amplitude  $H^{(n)}$  vs. bead position  $n$ .**

We observe that, for a given frequency  $\nu$ , the amplitude  $H^{(n)}$  does not vanish far away from the heat source (large  $n$ ). The result holds true in the frequency range explored experimentally and we measure that the limit  $H^{(\infty)}$  decreases when the frequency  $\nu$  is increased (Fig. 9).



**Fig. 9. Asymptotic amplitude  $H^{(\infty)}$  and  $k'$  vs.  $\nu$ .**

We observe experimentally that the asymptotic amplitude  $H^{(\infty)}$  decreases, according to equation (4), when the frequency  $\nu$  is increased. In addition,  $k' = \Re(k)$  obeys equation (5), which makes possible to characterize the thermal conduction along the chain.

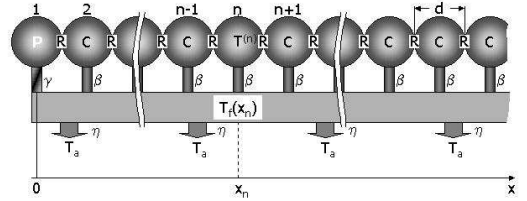
We observe a good agreement of the experimental data (Fig. 9) with

$$H^{(\infty)}(\nu) \sim \frac{1}{\sqrt{1 + \left(\frac{2\pi\nu}{\omega_c}\right)^4}} \quad (4)$$

for  $\omega_c = (0.8 \pm 0.01) 10^{-3} \text{ rad s}^{-1}$  where  $\omega_c^{-1} \equiv C\sqrt{\beta\gamma}$  from the model [see appendix, equation (18)]. In addition, we expect  $k'$  to be the real part of  $k$ , defined by

$$\cosh(k) = \left[1 + \frac{1}{\omega_0\tau_c}\right] + j\frac{2\pi\nu}{\omega_0}. \quad (5)$$

where  $\omega_0^{-1} \equiv RC/2$  and  $\tau_c \equiv \beta C$ . From equation (5) and the experimental data, we get  $\omega_0\tau_c = 1.87 \pm 0.03$  and  $\omega_0 = (7.6 \pm 0.1) 10^{-3} \text{ rad s}^{-1}$ . Note that the first quantity,  $\omega_0\tau_c \equiv 2\beta/R$ , is the ratio of the resistance  $\beta$  associated to the energy losses to the resistance  $R$  of the bead-bead contacts whereas  $\omega_0 \equiv 2/(RC)$  characterizes the transport of heat along the chain alone.



**Fig. 10. Sketch of the experimental situation.**

### 3.1.3 Quantitative analysis

In this section, we shall compare the experimental value of the contact resistance, which we can extract from the experimental  $\omega_0$ , with a theoretical modeling due to G. K. Batchelor and R. W. O'Brien [2].

First, one can find in the litterature the heat capacity  $c_s = 0.5 \text{ J g}^{-1} \text{ K}^{-1}$  and the density  $\rho_s = 8 \text{ g cm}^{-3}$  of steel (AISI 304), which lead to  $C = 2.1 \text{ J K}^{-1}$  for  $d = 1 \text{ cm}$ . Thus, from the experimental value of  $\omega_0$ , one obtains the experimental value,  $R_{exp} = (126 \pm 1) \text{ K W}^{-1}$ , of the resistance of the bead-bead contact.

Second, we can consider theoretically the heat flux  $\Psi \equiv (T^{(n)} - T^{(n+1)})/R$  from bead  $n$  to bead  $(n+1)$ . According to the Hertz theory, due to the axial applied force  $F$ , the radius  $\rho$  of the contact region is given by [8]:

$$\rho = F^{1/3} \left[ \frac{3(1 - \sigma^2)d}{8E} \right]^{1/3} \quad (6)$$

where  $E = 2 \cdot 10^{11} \text{ Pa}$  is the Young modulus and  $\sigma = 0.29$  the Poisson ratio of the material the beads are made of. In our experimental conditions ( $F = 100 \text{ N}$ ), we find

$\rho = 0.12$  mm. In addition, the heat conductivity of steel,  $k_s = 16.2$  W m<sup>-1</sup> K<sup>-1</sup>, is large compared the thermal conductivity of air,  $k_a = 0.025$  W m<sup>-1</sup> K<sup>-1</sup>. Denoting  $\zeta = k_s/k_a$  and  $\xi = 2\zeta\rho/d$  (in our experimental conditions,  $\zeta \simeq 650$  and, from the radius  $\rho$  of the contact region,  $\xi \simeq 15.6$ ), we can write

$$\frac{2\Psi}{\pi k_a d(T^{(n)} - T^{(n+1)})} = \mathcal{H}_0(\zeta) + \mathcal{H}_c(\xi) + \Delta\mathcal{H}_m(\xi). \quad (7)$$

In equation (7),  $\mathcal{H}_0(\zeta)$  stands for the dimensionless heat-flux across the air layer in the case of a point-like contact. Provided that  $\zeta \gg 1$ ,

$$\mathcal{H}_0(\zeta) = 2 \log_e \left( \frac{k_s}{k_a} \right) + A - 3.9 \quad (8)$$

where  $A$  is a constant of the order of unity which depends on the outer field conditions and is independent of  $\zeta$ . The second contribution  $\mathcal{H}_c(\xi)$  in equation (7) accounts for the dimensionless heat-flux across the contact circle of finite radius  $\rho$ . In the limit  $\xi \gg 1$ ,  $\mathcal{H}_c(\xi) \sim 2\xi/\pi$ . We point out that, from [2],  $\mathcal{H}_c(\xi)$  obeys this asymptotic behavior typically for  $\xi > 10$ . Finally, the last contribution to the heat flux in equation (7),  $\Delta\mathcal{H}_m(\xi)$  accounts for the difference between the real flux across the air layer and the flux expected in the case of a point-like contact. In the limit  $\xi \gg 1$ ,  $\Delta\mathcal{H}_m(\xi) \sim -2 \log_e \xi$  but we note that this asymptotic behavior is expected for very large values of  $\xi$ . From the experimental value of  $\xi$ , we expect  $\Delta\mathcal{H}_m(\xi) \simeq -0.5$  [2]. Finally, from equation (7), we obtain the theoretical value  $R_{theo} = 2546/(18.5 + A)$  K W<sup>-1</sup> which agrees quantitatively with the experimental value  $R_{exp} \simeq 126$  K W<sup>-1</sup> for  $A \simeq 1.7$ , which is of the order of unity. Thus, we conclude from the excellent agreement (in spite of the uncertainty in the value of  $A$ ) between  $R_{exp}$  and  $R_{theo}$  that measurements of the frequency  $\omega_0$  provides reliable values of the contact resistance  $R$ .

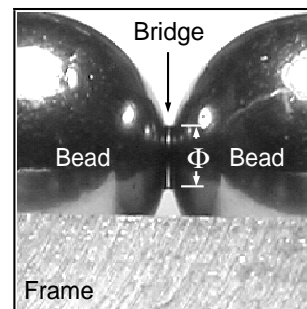
In the next section 3.2, we make use of the experimental setup for analyzing the effect of liquid bridges decorating the regions of contact between the beads.

### 3.2 Wet contacts

As already mentioned in the introduction, humidity has been experimentally proven to increase significantly the thermal conductivity of an assembly of glass beads as water decorates the contact points [12]. Our experimental setup is particularly suitable for studying the effects of liquid decorating the contact points: indeed, a change of the physical conditions in the contact region is expected not to affect any of the model parameters ( $\tau_c$ ,  $\tau_b$ ,  $\alpha$  as defined in the appendix) excepted the cut-off angular frequency  $\omega_0$ , which relates to the resistance of the contact. Thus, one can save experimental time and study the evolution of  $\omega_0$  alone by analyzing the response of the system at a given position  $n$  only.

#### 3.2.1 Experimental procedure

We shall report the evolution of the thermal resistance  $R$  of the bead-bead contact as a function of the size of the liquid bridges that decorate the contact regions. As an example, we choose to study the response of the system at the bead 4: we know from the study of the dry system that the amplitude of the temperature variations is large enough for giving significant results and that the order of the 'filter' is also large enough for providing accurate measurements of the cut-off frequency  $\omega_0$ . This choice is anyway in a large manner arbitrary.



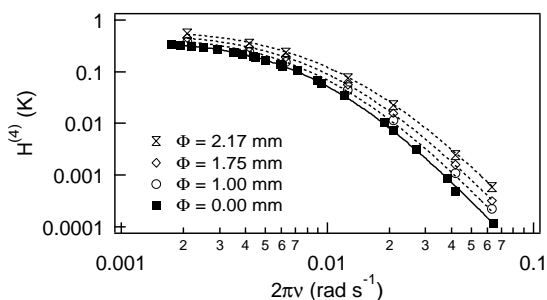
**Fig. 11. Photograph of a liquid bridge.**

We measure the diameter  $\Phi$  of the liquid bridge on photographs of the bead-bead contact-region ( $\Phi = 2.17$  mm, the lateral size of the photograph is 1 cm).

Nonvolatile liquid bridges (identical along the chain) are created by placing a given amount of silicon oil (Rhodorsil, 47V5000) in each of the bead-bead contact-region. We expect, from the large viscosity of the liquid ( $\nu = 5000$  cP), the temperature gradient not to induce any significant convective flow and the transport of heat to be only due to diffusion. We image the system from side (Fig. 11), which makes possible to measure the diameter,  $\Phi$ , of the liquid bridge (the variability along the chain remains less than 2%). We measure  $\phi$  before and after measurement of the thermal response of the system in order to make sure that the bridges were equilibrated during the experimental time.

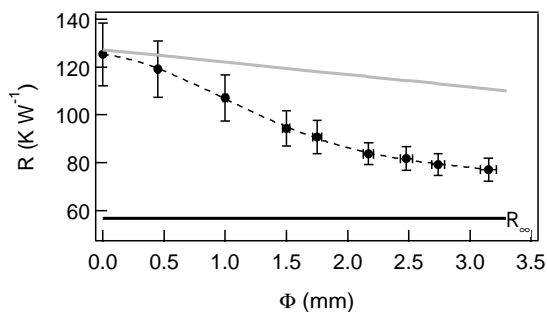
#### 3.2.2 Results

We report in figure 12, the amplitude  $H^{(4)}$  as a function of the frequency  $\nu$  for the dry system and three different diameters  $\Phi$ . The experimental results exhibit an excellent agreement with the theoretical expression (18) in which we use the experimental parameters,  $\tau_b = 318$  s,  $\tau_c = 246$  s,  $\alpha = 2289$  and  $\overline{P}\tau_b/C = 13.3$  K determined previously for the dry system and adjust only the value  $\omega_0$  of the cut-off frequency. From the experimental value of  $\omega_0$ , provided that the thermal inertia of the bridges remains negligible compared to that of the beads, we can easily determine



**Fig. 12. Amplitude  $H^{(4)}$  vs.  $2\pi\nu$ .**

The amplitude  $H^{(4)}$  is affected by the presence of liquid bridges in the contact regions between the grains: we observe that, at a given frequency  $\nu$ ,  $H^{(4)}$  increases with the diameter  $\Phi$  of the bridges. The presence of liquid leads to a decrease in the thermal resistance  $R$  of the contact. The experimental data (symbols) are successfully accounted by the theoretical description of the system [lines, from equation (18) with different values of  $\omega_0$ ].



**Fig. 13. Resistance  $R$  vs. bridge diameter  $\Phi$ .**

We observe that the thermal resistance of the contacts decreases when the diameter of the liquid bridge is increased. The dashed line is a guide for the eye while the horizontal black line corresponds to the value,  $R_\infty = 56.6 \text{ K W}^{-1}$ , estimated for beads totally immersed in oil [from equations (7) and (8) with  $A = 1.7$ ]. The grey line corresponds to equation (9).

the thermal resistance of the contact  $R$  which is found to decrease when the bridge diameter  $\phi$  is increased (Fig. 13).

The theoretical description of the thermal resistance associated with a bead-bead contact of finite size decorated by a liquid bridge is, to our knowledge, not yet available. The extension of the model briefly summarized in section 3.1.3 to this latter case appears to be difficult and we will limit the discussion of the experimental results to a few comments.

First, we can obtain an estimate of the thermal resistance  $R_\infty$ , one would measure for beads immersed in silicon oil, and compare this latter value to the minimum of  $R$  reported in figure 13. The thermal conductivity of the silicon oil  $k_{so} = 0.16 \text{ W m}^{-1} \text{ K}^{-1}$ . In this case,  $\zeta \simeq 101$  and  $\xi \simeq 2.4$  (for  $F = 100 \text{ N}$ ). Replacing  $k_a$  by  $k_{so}$  in equation (8), we get  $\mathcal{H}_0(\zeta) \simeq 5.33 + A$ , which accounts for the heat transport through the fluid layer around the contact ( $A$  is again a constant of the order of unity). Moreover, we know from reference [2] that, for a value of  $\xi$  as

small as 2.4, the value  $2\xi/\pi \simeq 1.5$  largely overestimates the real value of  $\mathcal{H}_c(\xi)$ , which is associated to the conduction through the contact circle of radius  $\rho$ , and that the correction  $\Delta\mathcal{H}_m(\xi)$  remains much smaller than unity. Thus, we can neglect  $\mathcal{H}_c(\xi)$  and  $\Delta\mathcal{H}_m(\xi)$  in equation (7), which leads to  $R_\infty \simeq 398/(5.33 + A) \text{ K W}^{-1}$ . If we assume that the value  $A = 1.7$  remains unchanged when the beads are immersed in air or in silicon oil, we get  $R_\infty \simeq 56.6 \text{ K W}^{-1}$  (Fig. 13). We observe that the resistance of the contact  $R$  approaches  $R_\infty$  when the diameter  $\Phi$  of the bridge is increased. Even if  $R$  remains about 30% larger than  $R_\infty$  for  $\Phi/d \simeq 0.3$ , we consider that the agreement between our experimental data and the theoretical estimate of  $R_\infty$  again reinforce the validity of our measurements. Moreover, we are taught by the model that, in the case of beads immersed in oil, the heat is mainly transferred through the liquid layer around the contact region and not through the circle of contact (indeed,  $\mathcal{H}_c(\xi) \ll \mathcal{H}_0(\zeta)$ ). We expect the conclusion to hold true in the case of a large liquid bridge decorating the contact region. This provides us with an important clue for understanding the smooth variation of  $R$  with the diameter  $\phi$  of the bridge.

We could expect from equations (7) and (8) and from the ratio air- to oil-conductivity,  $R$  to suddenly decrease when the liquid is introduced in the contact region. However, we observe experimentally that  $R$  decreases almost linearly when  $\Phi$  is increased. This feature may be qualitatively understood as follows: in the case of dry beads, about half of the heat is transferred through the real bead-bead contact, the second half passing through the air gap around the contact in spite of the small radius  $\rho$  of the contact circle. Think now about what would happen, if the radius  $\rho$  was slightly increased by  $\delta\rho$ . We would mainly expect  $\mathcal{H}_c(\xi)$  to increase linearly with  $\delta\rho$  as  $\mathcal{H}_c(\xi) \propto \rho$ ,  $\Delta\mathcal{H}_m(\xi)$  to remain negligible, and  $\mathcal{H}_0(\zeta)$  not to change. Note that to increase  $\rho$  is thus equivalent to decorate the contact with a steel bridge of diameter  $\Phi = 2(\rho + \delta\rho)$ . The corresponding variation  $\delta R$  of the resistance would then be given by  $\delta R = -R\delta\rho/\rho$  {from equation (7),  $\delta[\log_e(R)] = -\delta[\log_e(\mathcal{H}_c)]$ }. Thus, introduction in the contact region a small quantity of oil, which conductivity is smaller than that of steel, is expected to only continuously decrease the resistance of the contact and not to produce any discontinuous change in  $R$ . If we assume that the addition of oil produces the same effect as the addition of steel excepted that the transport of heat occurs in oil instead of in steel, we can roughly estimate

$$\frac{\delta R}{R} = -\frac{k_{so}}{k_s} \frac{\Phi - 2\rho}{2\rho} \quad (9)$$

which leads to  $\frac{dR}{d\Phi} \simeq -5.2 \text{ K W}^{-1} \text{ mm}^{-1}$ . As the presence of the liquid bridge modifies also the temperature field in the bead, leading to a spreading of the temperature-field lines and, thus, to reduced thermal resistance. The slope given by equation (9) is expected to overestimate the experimental value for small  $\Phi$ , which is observed experimentally (Fig.13). Obviously, due to assumptions which remain open to criticism, this quantitative estimate must be considered with caution. However, we consider that



the reasoning presented above is qualitatively correct for explaining the smooth dependancy of the resistance  $R$  on the diameter  $\Phi$ , when the contact region is decorated by a liquid bridge.

## 4 Conclusions

First, we would like to point out several flaws of our experimental situation; due to long thermal times (especially  $2\pi/\omega_0 \simeq 15$  mn) the experimental times are long [for instance, more than 10 hours are needed for measuring  $H^{(n)}(\nu)$  at five relevant frequencies  $\nu$ ]; the study of  $R$  as a function of the applied force  $F$  or of the nature of the surrounding gas, which is in principle possible, would require a huge experimental effort. Indeed, as at least the times  $\tau_b$  and  $\tau_c$  would depend on  $F$  or on the nature of the surrounding gas, characterization of the whole experimental setup would be necessary. The analysis could not be limited to the response of the system at a given position  $n$  in the chain, which would increase significantly the experimental time. Note however that the behavior of  $H^{(n)}(\nu)$  as a function of  $n$  at one given frequency  $\nu$  would be, in principle, enough for determining  $\omega_0$ . Such studies remain to be done although we already attempted, before drawing our conclusions, to measure the resistance of the contacts in air, nitrogen, carbon dioxide and helium and obtained sensible qualitative-results. However, these studies were limited to the thermal response at a given position  $n$  as a function of the frequency  $\nu$ , which unfortunately turned out not to be sufficient to determine  $R$  afterwards.

In conclusion, we have shown that the study of the heat transport along a chain of beads is a pertinent way to access the thermal properties of the contact. The experimental method is applied successfully to the case of dry contacts in the sense that the measured resistance  $R$  can be accounted by a classical model by Batchelor *et al* [2]. We made use of the experimental setup for studying the effect of liquid bridges decorating the contact regions and observed a smooth decrease in the thermal resistance when the diameter of the bridge is increased. Even if a theoretical description of this latter experimental situation is not yet available, simple arguments, which extend the model of the dry contact, allow to understand, at least qualitatively, the experimental results. This work provides important clues for understanding the transport of heat in partially wet granular materials.

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## Appendix

### Modeling of the experimental situation

We observe experimentally that the temperature oscillation at a given frequency  $\nu$  decreases exponentially when

$n$  increases and tends to a finite value for large values of  $n$ . These experimental observations are accounted by the following modeling of the experimental system (Fig. 10).

The thermal conductivity of the chain can be accounted as follows : we consider each bead as a thermal mass,  $C$ , in contact with its two neighbors (excepted bead 1) through identical thermal resistors,  $R$ . We assume that the thermal losses through the PTFE plates, which are used to align the beads, can be described as the effect of a single thermal resistor,  $\beta$ , which connects locally each bead to the remaining part of the experimental setup. Let us denote  $T^{(n)}(t)$  the temperature of the bead  $n$  at time  $t$ . We can thus write, taking  $\omega_0 \equiv 2/(RC)$  and  $\tau_c \equiv \beta C$ ,

$$\frac{\partial T^{(n)}}{\partial t} = \frac{\omega_0}{2} \left[ T^{(n+1)} + T^{(n-1)} - 2T^{(n)} \right] - \frac{1}{\tau_c} \left[ T^{(n)} - T_f(x_n) \right] \quad (n > 1) \quad (10)$$

where  $T_f(x_n)$  denotes the temperature of the experimental setup at the position  $x_n \equiv (n-1)d$  of the bead  $n$  (diameter  $d$ ). We will assume that the chain is long enough to be considered as semi-infinite: the boundary condition far away from the heat source then only writes  $T^{(n+1)} = T^{(n)}$ , ( $n \rightarrow \infty$ ). We will further assume that the contact between the first bead with the metallic part, that constrains the chain along its axis, can be described by a simple thermal resistor,  $\gamma$ . The first bead is heated by injecting the heating power  $P(t)$  so that the energy balance leads to, taking  $\tau_b \equiv \gamma C$ ,

$$\frac{\partial T^{(1)}}{\partial t} = \frac{\omega_0}{2} \left[ T^{(2)} - T^{(1)} \right] - \frac{1}{\tau_b} \left[ T^{(1)} - T_f(0) \right] + \frac{P}{C}. \quad (11)$$

Let us now assume that the experimental setup, apart from the PTFE plates and the chain, behaves like a semi-infinite rod (diffusion coefficient  $D$ ) in contact with the outside atmosphere (at temperature  $T_a$ ) through the conductance per unit length  $1/\eta$ . Furthermore, we assume that the thermal contact between the beads and the rod along the chain is bad enough for the temperature of the rod not to be affected by the corresponding heat sources (*i.e.*  $\beta$  is large and the diffusion in the rod is fast). As a consequence, we can write :

$$\frac{\partial T_f}{\partial t} = D\Delta T_f - \frac{1}{\tau_l} (T_f - T_a). \quad (12)$$

where  $\tau_l = \eta C_f$  is the characteristic time associated with the thermal losses to the atmosphere ( $C_f$  is here the heat capacity per unit length of the metallic frame). We will consider that the temperature profile  $T_f(x, t)$  only results from energy injection at  $x = 0$  from bead 1 (we thus assume that  $\gamma \ll \beta$ ). Writing the energy balance at the contact between bead 1 and the rod leads to :

$$\frac{T^{(1)} - T_f(0)}{\gamma} = -DS\nabla T_f(0) \quad (13)$$

where  $S$  stands for the cross section of the rod.

After having written, the general set of equations (10) and (12) and the boundary conditions (11) and (13), we consider the harmonic response of the experimental setup as a function of the frequency  $\nu = \omega/(2\pi)$  by writing :

$$T_f(x, t) = T_0 \exp(j\omega t - k_f x), \quad (14)$$

$$T^{(n)}(t) = T_0^{(n)} \exp(j\omega t), \quad (15)$$

and

$$P(t) = \bar{P}(\omega) \exp(j\omega t). \quad (16)$$

From equation (12), we get  $k_f^2 = \frac{\eta + j\omega}{D}$  whereas the boundary condition (13) leads to  $T_0^{(1)} = T_0(1 + \alpha)$  with  $\alpha \equiv \gamma D S k_f$ . In addition, equation (10) imposes :

$$T_0^{(n)} = T_0^{(0)} \exp[-(n-1)k] \quad (17)$$

$$+ T_0 \frac{\exp[-(n-1)k_f d]}{1 + \omega_0 \tau_c [1 - \cosh(k_f d)] + j\omega \tau_c}$$

with  $\cosh(k) = [1 + \frac{1}{\omega_0 \tau_c}] + j \frac{\omega}{\omega_0}$ . The constants  $T_0^{(0)}$  and  $T_0$  are obtained as functions of  $\bar{P}$  by using the second boundary condition (11).

In the limit  $|k_f d| \ll 1$ , which is well satisfied in our experimental conditions, equation (17) reduces to

$$\frac{C T_0^{(n)}}{\bar{P} \tau_b} = \left\{ 1 + [\alpha + j(1 + \alpha)\omega \tau_c] \exp[-(n-1)k] \right\} /$$

$$\left\{ (1 + j\omega \tau_c) [\alpha + j(1 + \alpha)\omega \tau_b] \right. \quad (18)$$

$$\left. + \frac{1}{2} \omega_0 \tau_b [1 - \exp(-k)] [\alpha + j(1 + \alpha)\omega \tau_c] \right\}$$

We point out that the diffusion coefficient in the rod,  $D$ , only appears, in this final expression proposed for  $|T_0^{(n)}|$ , in the coefficient  $\alpha$ . We are in the limit  $|nk_f d| \ll 1$ , which leads to  $|k_f d| \ll 1$  when we consider the limit  $n \gg 1$  (the diffusion in the rod is fast) but, even in this limit,  $\alpha \equiv \gamma D S k_f$  can remain finite. Note that, from equation (18), the amplitude  $T_0^{(n)}$  of the temperature oscillations decreases exponentially when  $n$  is increased and reaches, for  $n$  large, the value,  $T_0^{(\infty)}$  which magnitude remains finite as observed experimentally [Fig. 9, Eq. (4)].

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