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Aging in humid granular media

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Aging behavior is an important effect in the friction properties of solid surfaces. In this paper we investigate the temporal evolution of the static properties of a granular medium by studying the aging over time of the maximum stability angle of submillimetric glass beads. We report the effect of several parameters on these aging properties, such as the wear on the beads, the stress during the resting period, and the humidity content of the atmosphere. Aging effects in an ethanol atmosphere are also studied. These experimental results are discussed at the end of the paper.

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I. INTRODUCTION

Granular media have many interesting and unusual properties [1]. The intrinsic dissipative nature of the interactions between the constituent macroscopic particles sets granular matter apart from conventional gases, liquids or solids. One of the most interesting phenomena in granular systems is the transition from a static equilibrium to a granular flow. Contrary to ordinary fluids, they can remain static even with an inclined free surface. But when the angle of the surface with the horizontal exceeds some threshold value θ_m, the pile cannot sustain the steep surface and starts to flow until its angle relaxes under a given angle of repose θ_r.

The friction properties of grains play an important role in this transition from static equilibrium to flow [2]. Indeed, the phenomenological laws of static friction formulated by Coulomb in 1773 identified the existence of a definite avalanche angle in a granular media to a friction coefficient: $\tan\theta_m = \mu$. Subsequent works have emphasized the similarities between solid-solid friction and friction in granular media. On the one hand, detailed studies in solid-solid friction have brought into evidence temporal evolution in friction phenomena between solids. In a number of materials the static friction coefficient increases logarithmically with the contact time, whereas the dynamic friction coefficient decreases as the logarithm of the slipping velocity [3]. These temporal evolutions have been shown to be an effect of the evolution over time of contacts between surfaces asperities—the aging of contacts, and result in stick-slip behavior at low-velocity motion [4]. Recently, Nasuno *et al.* [5], and Geminard *et al.* [6] have reported measurements of stick-slip transition on granular layers of spherical glass beads. A similar study has been made by Lubert and de Ryck [7] between silica gel grains in an annular geometry. On the other hand, the temporal evolution of friction properties in a granular material has been studied by Bocquet *et al.* [8], who report a slow increase of the maximum stability angle of submillimetric glass beads with the time of rest of the granular heap. Nevertheless, this temporal evolution is observed only in a humid atmosphere. This important effect of humidity in the aging properties of friction has also been reported by Dieterich and Conrad in rock-to-rock friction [9], and Crassous *et al.* on paper-to-paper and glass-to-glass friction [10]. This shows that a small amount of liquid in the atmosphere can drastically modify the friction properties of solids.

A number of recent experiments have studied the effect of a small amount of liquid on the maximum stability angle in a granular material. Barabasi and co-workers [11–14] as well as Mason *et al.* [15], and Halsey and Levine [16] have studied the influence of a small quantity of nonvolatile liquid added to a granular medium, and shown an important increase of its maximum stability angle. Using water vapor and alcane vapor, Fraysse *et al.* [17] have reported a significant increase in the maximum stability angle with the vapor content of the surrounding atmosphere.

These experiments do not explicitly incorporate time as a parameter for cohesion effects in granular media as studied by Bocquet *et al.* [8]. More recently, Ovarlez *et al.* [18] have shown a humidity-induced aging effect on the dynamical behavior of a granular column pushed vertically. A similar effect has been reported by D'Anna [19] in frictional properties of granular in a Couette geometry. In this paper, we investigate more precisely the temporal effects on the avalanche angle of submillimetric glass beads. For this purpose, we use a rotating drum and perform experiments at a controlled temperature and humidity. We have also performed experiments in ethanol vapor. We analyze in detail the effect of parameters such as the tangential stress during the resting period and the wear of the beads on the temporal dependence of the avalanche angle. We show that this temporal evolution results from the increase of the cohesion force between grains when the beads are in a vapor atmosphere.

II. EXPERIMENTAL SYSTEM

A. The setup

The experimental setup is described in Fig. 1. The granular material is inserted in a cylindrical drum of stainless steel with an inner diameter and a length of 10 cm. This geometry reduces the influence of the wall on the stability angle (the

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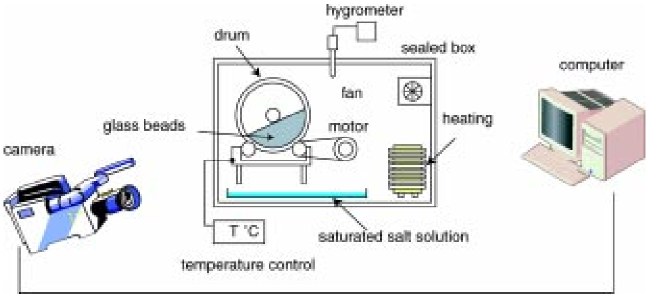

FIG. 1. Experimental setup.

typical length scale on which the wall affects the stability angle is on the order of ten bead diameters [20]). The lateral faces of the cylinder are made of glass with an opening at the center sufficiently large to allow vapor exchange with the outer atmosphere. The drum is partially filled with glass beads. In all the experiments we have used the same height of glass beads (4 cm). In order to keep the relative humidity of the atmosphere constant during the experiments, the setup is placed in a sealed lucite box. The temperature in the box is controlled by a heating resistor, a fan enclosed in the box, and a temperature controller. The heating resistor is a THERMOCOAX heating element connected to a digital temperature controller (REX F400, TCSA Dardilly, France). The temperature measurement is done with a Pt100 inserted in the box. The temperature is fixed at (29 ± 0.2) °C.

The cylinder can rotate around its horizontal axis at a constant speed Ω controlled with a dc motor. The rotation speed can be varied from ~ 0.01 to 60 rpm. One of the lateral faces of the cylinder is monitored by a video camera connected to a computer. The maximum slope of the surface is then directly measured with SCION Imaging Software. The error on the measurements is typically $\pm 1°$. The main error is due to the lack of flatness of the heap surface. This error is smaller than the dispersion of the measurements, which are due to the small differences in the preparation of the samples, small drifts, and unwanted vibrations. We report the avalanche angle measured on the surface of the glass windows on the lateral faces of the cylinder. This angle is always greater than the avalanche angle in the middle of the drum. The difference between these two angles (typically 2°) does not change with the waiting time.

The glass beads used in this study are industrially used for sandblasting (see Fig. 2) and are sold by Matrasur (Marcoussis, France). These glass beads are smooth and have a roughly spherical geometry.

In all the forthcoming experiments, the diameter of the beads is smaller than 0.3 mm. We have found that the temporal effects become small for glass beads larger than 0.3 mm. We used three samples of glass beads (hereafter A,B,C), with sizes calibrated by sifting (see Table I).

B. Humidity control

Two parameters are used to quantify the amount of vapor water in the atmosphere. These are the partial pressure of water p_{vap}, or the relative humidity $H_{\mathrm{R}}=p_{\mathrm{vap}}/p_{\mathrm{sat}}$, defined as the partial pressure of water divided by the saturating vapor pressure p_{sat}. The relative humidity H_R is often expressed as a percentage: $H_{\mathrm{R}}(\%)=H_{\mathrm{R}}\times 100$. The relative humidity is a more pertinent parameter since it is related to the thermodynamical properties of the liquid-vapor equilibrium. Indeed, the undersaturation $\Delta\mu=\mu_{\mathrm{vap}}-\mu_{\mathrm{sat}}$, defined as the difference between the chemical potential of the vapor μ_{vap} and the chemical potential at the liquid-vapor equilibrium μ_{sat}, can be expressed as [22]

$$\Delta\mu=\mu_{\mathrm{vap}}-\mu_{\mathrm{sat}}=kT\ln\left(\frac{p_{\mathrm{vap}}}{p_{\mathrm{sat}}}\right)=kT\ln(H_R), \tag{1}$$

where k is Boltzmann's constant and T is absolute temperature.

In the literature, different methods for the control of the relative humidity are described. Bocquet *et al.* [8] used the method of the controlled leak between a water container and the chamber containing the experiment. This method requires an adjustable control of the flow of vapor between the two containers and an accurate hygrometer. Fraysse *et al.* [17] used a more complicated method consisting in: (i) putting the beads under vacuum, and (ii) injecting a controlled amount of water (undersaturated vapor). This method is a versatile method that can be implemented with any volatile liquid, resulting in a shorter equilibrium time. In our experiments, we used another method to control the atmosphere. The relative humidity is kept constant by using an aqueous solution saturated with inorganic salts. A large beaker of saturated salt solution is put in the box. Each salt fixes the relative humidity of the surrounding atmosphere at a given temperature. The purity of the used salt exceeds 99% (in fact, a small amount of impurity does not induce a significant drift of the humidity). Table II shows that temperature has little influence on the relative humidity. The relative humidity H_{R} is measured during the experiments by a capacitive hygrometer with a precision of $\pm 2\%$.

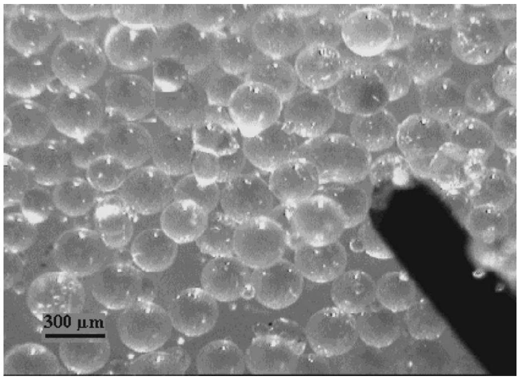

FIG. 2. Picture of a sample of glass beads with an optical microscope.

TABLE I. Size of the beads in the different samples.

Sample	Min. size (μm)	Max. size (μm)
A	200	250
B	125	160
C	0	50

TABLE II. Relative humidity of the air surrounding a saturated salt solution for different salts at different temperatures (after the AFNOR norm X15-111 [21]).

Saturated salt solution	H_R (%)									
Temperature (°C)	5	10	15	20	25	30	35	40	50	60
Lithium chloride (LiCl)	14	14	13	12	12	12	12	11	11	10
Potassium acetate (KCH_3CO_2)		21	21	22	22	22	21	20		
Magnesium chloride ($MgCl_2$)	35	34	34	33	33	33	33	32	31	30
Potassium carbonate (K_2CO_3)		47	44	44	43	43	43	42		36
Sodium chloride (NaCl)	76	76	76	76	75	75	75	75	75	76

C. Procedure

Our purpose is to study the aging effects on the avalanche angle of the granular medium. Therefore, we measure the avalanche angle of the granular heap after a waiting time t_w, the waiting time being defined as the time elapsed between the onset of an avalanche and the last overall motion of the grains before the avalanche. The procedure for measuring the avalanche angle after a resting period of length t_w is the following.

(1) First of all, the beads are introduced in the drum and rotated at a low speed (0.01 rpm) for 48 h in the controlled environment. After this preparation, we estimate that the humidity and temperature are homogeneous throughout the bulk of the granular medium.

(2) The drum is rotated for about one minute at an angular velocity of about 20 rpm, in order to create an overall motion in the granular medium.

(3) At time $t=0$ this quick rotation is stopped. We check that the free surface of the heap obtained is flat enough so that the value of the maximum slope can be measured within 2°. The position of the drum is adjusted so that the angle of the free surface of the heap with a horizontal plane has a well-defined value. We call this angle the reference angle θ_{ref}. The reference angle is always smaller than the maximum stability angle of the heap.

(4) The granular heap stays at rest until $t \simeq t_w$. The drum is then set in rotation at very low speed, and the angle of the free surface of the heap is recorded. The value of t_w and of the maximum stability angle are determined from the last image before the onset of the avalanche. Other experiments are then run from step 2.

We have probed a large range of waiting times starting from $t_w = 10$ s to $t_w = 3$ days. Experiments with different waiting times are performed in random order.

In the case of high humidity or large waiting time, the granular heap can be so cohesive that the heap can clump. In this case, it is not possible to obtain a flat surface when the heap breaks. In the following work, we have restricted our study to granular heaps with low cohesion forces so that a flat surface can be obtained.

III. HUMIDITY-INDUCED AGING

We first studied the influence of the resting time t_w on sample *A* in an atmosphere of relative humidity $H_R = 43\%$. The avalanche angle θ_m is plotted as a function of t_w in Fig. 3. The data clearly show an increase of the avalanche angle with the waiting time. This variation is large; the maximum stability angle is about 30° for a short waiting time and rises up to 55° after three waiting days. Even for longer waiting times, no saturation of the maximum stability angle was observed.

As a first approach, we may compare this temporal behavior to the aging properties of the static friction coefficient in normal solid to friction. In a number of materials, the static friction coefficient μ is found to increase logarithmically with the waiting time during which the two solids have remained at rest with respect to each other: $\mu = a + b \log_{10} t_w$. Following the law of Coulomb for the avalanche angle of the granular heap, one may expect a slow temporal growth for $\tan \theta_m$, for instance, as $\tan \theta_m = a + b \log_{10}(t_w)$. However, this crude analogy is not adequate to describe the aging behavior observed here, since it leads to a saturation value of the maximum stability angle equal to 90°. Indeed, in a purely frictional granular material, any negative normal stress applied on the surface of the heap should destroy the heap. This was not observed in our experiments. With large humidities ($H_R(\%) > 80\%$) and long waiting times, we observed a clumping of the granular material. In such a situation the surface of the heap reaches $\theta > 90°$. Clearly, cohesion forces retain the grains of the material together so that the heap can

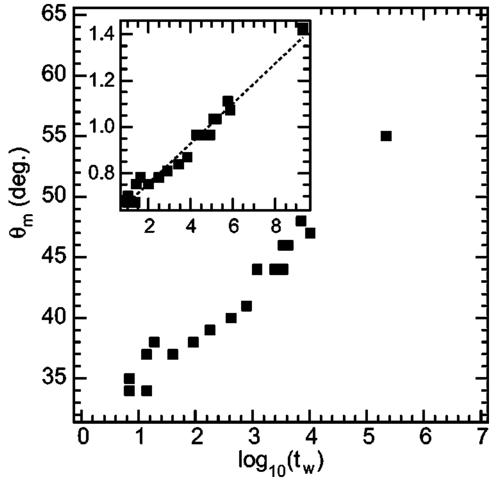

FIG. 3. The maximum stability angle θ_m as a function of $\log_{10} t_w$ for a sample of glass beads with a diameter between 200 and 250 μm. In these experiments the value of the reference angle is $\theta_{\mathrm{ref}} = 28°$ and the relative humidity is $H_R(\%) = 43\%$. In the inset, $\tan \theta_m$ is plotted as a function of $\log_{10} t_w / \cos \theta_m$. The dashed line is the best linear fit of the data.

stay stuck on the upper part of the drum due to this cohesive stress.

In order to take into account these adhesive forces, one may use a phenomenological extension of the Coulomb analysis. An equivalent formulation is to introduce the Mohr-Coulomb criterion that relates the normal stress σ across some plane interior to the sandpile to the maximum tangential stress τ possible without failure of the pile. In the absence of cohesive stress, a Mohr-Coulomb criterion for a granular medium is $\tau = \mu\sigma$. A cohesive stress can be taken into account by a phenomenological criterion [23]

$$\tau = c + \mu\sigma_n , \tag{2}$$

where σ_n is the normal stress introduced by external fields, and c the normal stress due to attractive interaction between grains. The maximum stability angle may then be obtained by considering the equilibrium of a layer of thickness D along the surface of the heap. The modified Mohr-Coulomb criterion (2) leads to a failure where

$$\rho g D \sin\theta = c + \mu\rho g D \cos\theta,$$

$$\tan\theta = \mu + \frac{c}{\rho g D \cos\theta}, \tag{3}$$

where ρ is the specific weight of the pile. This phenomenological description of the effect of cohesion forces on the stability of the sand pile explains the fact that the free surface of the pile can reach a vertical position. This occurs when the cohesive stress c is strong enough to sustain the weight $\rho g D$ of the layer. Halsey and Levine have conducted this analysis in more detail, and have predicted that in presence of cohesion forces the failure of the heap should occur at its bottom [16]. Also, the dependence of the avalanche angle with cohesion forces described in Eq. (3) has been probed recently by Forsyth *et al.* [24], who have used well-controlled magnetic adhesion forces between grains.

In the inset of Fig. 3 we have plotted the tangent of θ_m as a function of $\log_{10}t_w/\cos\theta_m$. The data are correctly fitted by the straight line such as

$$\tan\theta_m = \tan\theta_0 + \alpha\frac{\log_{10}t_w}{\cos\theta_m}, \tag{4}$$

where θ_0 and α are two adjustable coefficients. According to Eq. (3), this result means that there is a cohesion stress in the granular media which increases slowly in time according to: $c(t_w) = c_0\log_{10}(t_w)$. In the following, we characterize the influence of various parameters on the aging behavior by plotting the experimental data in this representation. We focus mainly on the coefficient α, which measures the the amplitude of the aging behavior of friction in the granular medium.

IV. PARAMETERS INFLUENCING THE AGING

A. The wear

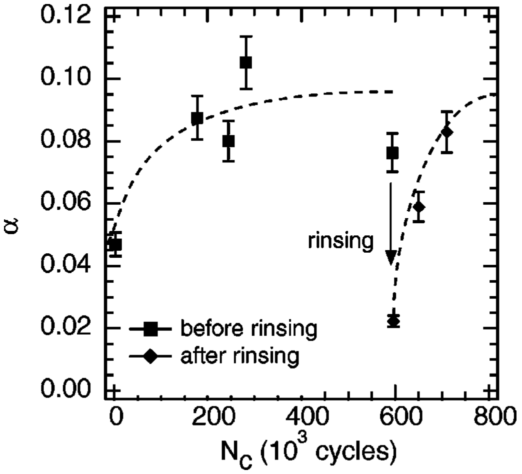

FIG. 4. Influence of the wear on the amplitude α of the aging behavior. N_c is the number of revolutions of the drum after which aging is measured. An arrow indicates that after this experiment the beads have been rinsed as explained in the text. The dashed lines are only guides to the eye.

We have studied the influence of the preparation of the sample of glass beads on their aging behavior. For this purpose we have taken 2.5 kg of glass beads directly as bought from Matrasur, and sifted them to calibrate their diameter in the 125–160-μm range (sample B). Then we have performed a first set of measurements of the aging coefficient α of this sample. After this first measurement, we have worn down the beads by rotating the drum at a higher speed (60 rpm) during a number of revolutions of the drum $N_c = N_1$. Then we have produced a set of avalanches to measure the new value of the aging coefficient. Then we have worn down the beads again during N_2 revolutions. The total number of revolutions of the beads is now $N_c = N_1 + N_2$. The evolution of the aging coefficient α as a function of the total numbers of revolution N_c is plotted on Fig. 4. This curve shows a slow increase of α with N_c and a saturation of the aging coefficient. After these five measurements, the glass beads were taken out of the drum and rinsed in distilled water several times. After the first rinse, the remaining water was turbid. We rinsed the beads until the rinsing water is no longer turbid. This suppresses all the small particles with a diameter typically smaller than 1 μm. The glass beads were dried, and the amplitude of aging measured again. In Fig. 4, the rinsing operation is symbolized by the vertical arrow. Just after rinsing, the aging coefficient has a very small value, even lower than the one obtained for the glass beads as coming from the supplier. We then have worn down the sample again, and we observed that the aging coefficient increases and comes back to the values obtained before the rinsing operation.

Therefore, the amplitude of the aging of the beads depends on their wear, but worn-down beads can be regenerated by rinsing. This experiment reveals that the effect of wear is mainly due to the production of glass dust, which plays an important role in the aging phenomenon. Also, the effect of wear is important for ‘‘clean’’ beads but tends to saturate after a certain number of revolutions of the drum. As a result, we subsequently have used worn-down beads to study the influence of other parameters on the aging behavior of the beads.

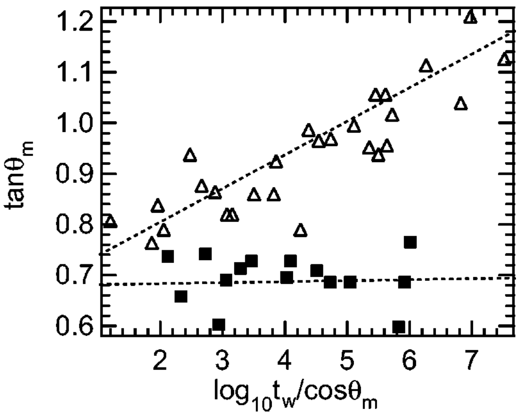

FIG. 5. Temporal evolution of the maximum stability angle for different values of the reference angle θ_{ref} at which the granular heap remains at rest. Filled squares : $\theta_{\mathrm{ref}}=0°$; open triangles : $\theta_{\mathrm{ref}}=28°$

B. Influence of the reference angle

We also have studied the influence of the reference angle θ_{ref} on the aging behavior of friction in glass beads. Figure 5 compares the evolution of the avalanche angle for a sample of glass beads which ages with a horizontal free surface ($\theta_{\mathrm{ref}}=0°$) and a sample of glass beads which ages at a reference angle close to the first avalanche angle measured at a short time: $\theta_{\mathrm{ref}}=28°$. The relative humidity in this experiment is $H_{\mathrm{R}}=43\%$. The beads' size is the same as in the previous experiments (sample B). One can see in Fig. 5 that aging is much smaller for beads that rest horizontally than for beads that rest at an angle close to the avalanche angle.

We believe that this effect is due to very small displacements occurring in the bulk of the heap when it is raised from an horizontal position to the avalanche angle. The effect of these small displacements is to break cohesive contacts between grains. In their study of granular friction, Nasuno *et al.* [25] report that in a stick-slip regime, some very small displacements (creep) precede the rapid events. The same precursors also have been reported in solid friction by Baumberger *et al.* [26]. Those precursors change dramatically the history of the adhesive contacts by breaking old ones and reforming new ones. To induce such changes, their amplitude has to overcome the range of the cohesive forces. In our experiment such precursors are much more numerous and important when the heap is tilted from an horizontal position up to the avalanche angle, which explains the weak aging effect obtained in this case. We will discuss in the last section the expected range of the cohesion force between the grains. This range must be small because the precursors obtained in solid friction or in granular friction are about 1 μm at most.

C. Influence of the surrounding humidity

When the wear of the beads and the reference angle are conveniently chosen, the amplitude of the aging behavior is reproducible and the influence of the surrounding humidity can be studied. The influence of the humidity p_{vap}/p_{sat} on the aging coefficient of a sample A of glass beads is plotted in Fig. 6. The main results are

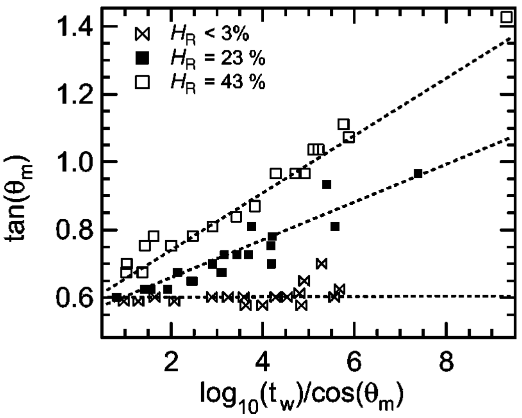

FIG. 6. Time evolution of the maximum stability angle θ_m of a 200 μm glass beads heap for various relative humidities.

(i) That no aging is observed in the dry case. It means that the aging behavior is not due to any slow compaction effect of the heap ([27]).

(ii) That the magnitude of the aging effect increases dramatically with humidity.

At this point, an important question concerns the physical origin of the slow increase of cohesion forces in an humid atmosphere revealed by the aging phenomenon. In order to check whether the influence of the water vapor is physical or chemical, we studied the influence of another condensable vapor: ethanol.

V. AGING IN AN ATMOSPHERE OF ETHANOL

A. Control of the partial pressure of ethanol

Unlike the case of water, there is no tabulated value of the influence of a salt dissolved in liquid ethanol on its equilibrium vapor pressure. Therefore, we first have designed a method to control the partial pressure of ethanol vapor, based on the one used to control water humidity (Figs. 7 and 8).

The experimental setup is described in Fig. 7. We measured the absolute pressure over a solution of ethanol saturated with different salts at different temperatures. The solution of ethanol is placed in a bath with a controlled temperature. The sensors then where thermalized at the temperature of the bath to prevent any condensation of ethanol vapor. Because ethanol is a polar liquid, it can dissolve some

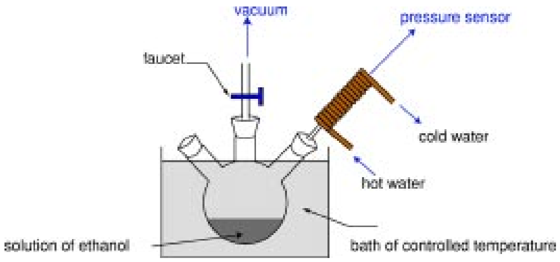

FIG. 7. (Color online) Schematic representation of the experimental setup used to calibrate the influence of an inorganic salt on the equilibrium pressure of ethanol vapor in the surrounding atmosphere.

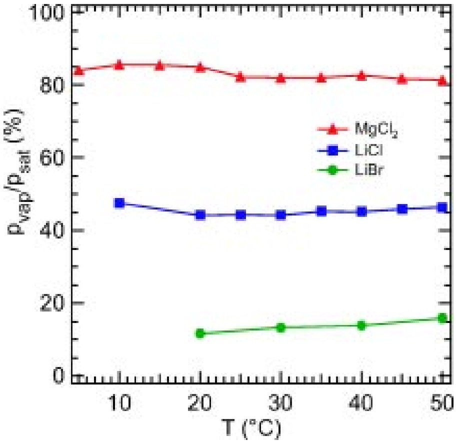

FIG. 8. (Color online) Evolution of the equilibrium humidity of ethanol for different saturated solutions as a function of temperature. The humidity is defined here as the ratio $p_{\mathrm{vap}}/p_{\mathrm{sat}}$ of the equilibrium pressure p_{vap} of ethanol vapor above the saturated solution to the saturating vapor pressure p_{sat} of ethanol vapor above pure liquid ethanol.

inorganic salts. In this study, we used $MgCl_2$, LiCl, and LiBr. As for the water solution, this modifies the chemical potential of ethanol in the liquid phase and, thus, the pressure of the vapor in equilibrium with this solution. Indeed, the partial pressure of ethanol strongly depends on the temperature [28], but we observed that the ratio of the partial pressure of the vapor at a given temperature, divided by the tabulated saturating pressure, does not really depend on the temperature. Typically, we obtained $p_{\mathrm{vap}}/p_{\mathrm{sat}}=0.84$ for $MgCl_2$, $p_{\mathrm{vap}}/p_{\mathrm{sat}}=0.44$ for LiCl, and $p_{\mathrm{vap}}/p_{\mathrm{sat}}=0.16$ for LiBr.

B. Aging in ethanol atmosphere

In an atmosphere of ethanol, we studied the evolution of the first avalanche angle of glass beads with their resting time. We have not found any aging behavior for glass beads of 200 μm diameter. Since the surface tension of ethanol is lower than the surface tension of water, we checked the effect of ethanol on a sample of smaller glass beads. Fig. 9 shows that aging can be observed for glass beads with a diameter comprised between 0 and 50 μm. On this plot, the data are more dispersed than for water vapor because temperature was not controlled in these experiments for safety reasons.

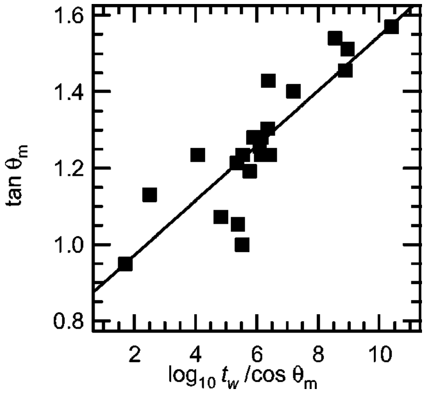

FIG. 9. $\tan\theta_m$ as a function of $\log_{10}t_w/\cos\theta_m$ for a sample of glass beads with a diameter between 0 and 50 μm. The temperature is not controlled in this experiment. A vapor pressure $p_{\mathrm{vap}}/p_{\mathrm{sat}}=0.84$ of ethanol is present in the atmosphere of the box. The straight line is the best linear fit of the data.

VI. DISCUSSION

These experiments on the aging behavior of friction in a granular heap of glass beads show that this aging is mainly due to a slow increase in time of cohesion forces inside the heap. As an order of magnitude, we find that the avalanche angle of a heap of 200 μm diameter glass beads may increase from 35° to 55° in two days in an ambient atmosphere of 40% relative humidity.

We have shown that this aging phenomenon is significantly increased when the beads are worn down so as to contain a certain amount of glass dust. This can be understood by the fact that glass dust increases the specific area of the granular medium and, thus, the total amount of liquid that can condense in it under otherwise fixed conditions. Also, the aging behavior is very sensitive to the tangential stress applied to the medium when it is at rest. In our experiments these effects are due to small displacements that occur in the medium when it tangentially is loaded and whose effect is to break old contacts and to create new ones. We do not know if it is an intrinsic property of granular friction, or if it comes from the fact that our granular medium is loaded only by its own weight. Finally, we have not found any saturation of this temporal evolution of the avalanche angle. We attribute this to the fact that when cohesion effects become important, clumping and cohesive structures appear in the bulk of the heap, and our experimental setup is not adapted to the measurement of a maximum stability angle.

A very important parameter influencing aging is the humidity content of the atmosphere. Bocquet *et al.* [8] have proposed a physical mechanism for this humidity-induced aging. It is based on the slow increase of the cohesion force in a granular medium due to the capillary condensation of small liquid bridges between the grains. It has been shown both experimentally [14] and theoretically [16] that the capillary forces created by liquid bridges can very significantly increase the maximum stability angle of a granular medium. Hornbaker *et al.* [11] have also shown by adding mineral oil to polystyrene beads—a chemically inert system—that the capillary forces induced by liquid bridges are strong enough to result in global clumping of the granular heap. In this approach, time does not appear as a parameter since the liquid is directly added to the granular medium. However, in the presence of a condensable vapor, the liquid bridges could condense in the medium with a slow kinetics. Direct measurements of capillary condensation of a liquid bridge between solid surfaces performed with surface forces apparatus [29,30] show that a thin film of vapor confined between wetting surfaces can be highly metastable, whereas the stable state is a liquid bridge. Restagno *et al.* [31] have evaluated the energy barrier for the condensation of such liquid bridges, and have shown that the activation energy grows like the power 3/2 of the distance between the solid surfaces, and may become very large. Bocquet *et al.* [8] then suggest a mechanism of thermally activated capillary condensation of liquid bridges between random asperities of the contact between grains. Assuming a random distribution of the asperities, they obtain a large distribution of activation energies, which result in a slow increase in time of the capillary force between grains.

The experiments reported here do not allow us to determine if the increase in cohesion forces in the heap of glass beads in a humid atmosphere is due to a purely physical mechanism, as proposed by Bocquet *et al.*, or a chemical mechanism, such as the sintering of glass beads due to a chemical reaction between of silica and water. We have observed an aging phenomenon in ethanol atmosphere, which is much less reactive with glass than water. However, aging in this case is observed only with beads with a diameter significantly smaller than the one used with water. In order to quantitatively compare the two systems, one can try to estimate by a dimensional argument the effect of capillary forces in the bulk of the heap. The capillary force between two beads of radius r is $2\pi\gamma_{LV}r$, and the number of bead contacts across a plane in the medium scales as $1/r^2$. Therefore, the cohesive stress due to capillary force should scale as γ_{LV}/r. On the other hand, the bulk density of the granular medium does not vary much with the size of the beads. Thus, assuming that the kinetics of condensation of liquid bridges is the same in an ethanol or a water atmosphere, one would expect similar effects for a ratio of beads radii of $\gamma_{\text{water}}/\gamma_{\text{ethanol}} = 2.8$. In our experiment, the beads' radii ratio in sample A and C is more than 5, and aging is observed in an ethanol atmosphere for a relative humidity much higher than that in a water atmosphere. Therefore, it is not possible to conclude that capillary forces only contribute to the aging phenomenon. In order to more precisely test the physical origin and the time dependence of the adhesion force between beads, this adhesion force should be measured directly between controlled surfaces and in a controlled atmosphere. The surface forces apparatus, which directly measures the adhesion force between two macroscopic surfaces, could be an ideal way to do these studies, since it has been already used to study the kinetics of growing of a liquid bridge of capillary condensation between ideal surfaces [32].

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