Mechanisms for Acoustic Absorption in Dry and Weakly Wet Granular Media

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The dissipation of an elastic wave in dry and wet glass bead packings is measured using multiple sound scattering. The interplay of a linear viscoelastic loss and a nonlinear frictional one is observed in dry media. The Mindlin model provides a qualitative description of the experiment, but fails to quantitatively account for the data due to grain roughness. In weakly wet media, we find that the dissipation is dominated by a linear viscous loss due to the liquid films trapped at the grain surface asperities. Adding more liquid enables us to form the capillary menisci but does not increase the energy loss.

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The attenuation of elastic wave in earth materials has been the subject of extensive investigations for many years in the fields of both seismology and rock physics [1–4]. The acoustic properties of porous granular materials such as sedimentary rocks, ocean sediments, and sintered bead packs are in general characterized by the properties of solid frame, the pore fluid if any, and frame-fluid interactions. Several mechanisms of intrinsic attenuation have been proposed, including the Coulomb frictional sliding between cracks and grain boundary contacts and the linear mechanism dominated by viscous dissipation of bulk fluid flow in partially or fully saturated rocks. In *room-dry* rocks containing small amount of volatiles, it is found that the attenuation is caused by another linear mechanism, due to a stress-induced diffusion of adsorbed monolayers of volatiles [3].

Noncohesive granular materials are composed of a dense random packing of macroscopic particles. Unlike consolidated porous materials, granular materials acquire basically its elasticity as a result of the applied stresses, forming the very inhomogeneous force network—a solid frame [5,6]. In the long-wavelength limit, the elastic wave propagation can be described by the effective medium approach based on the Hertz-Mindlin theory of contact at the grain level [7–9]. Similar to porous rocks, a small amount of liquid has a drastic effect on the mechanical properties of granular materials, such as the angle of avalanche and sound propagation [10]. Pilbeam and Vaisnys [11] have measured acoustic velocity and attenuation in dry and lubricated granular materials. They found that measured velocities in all the samples could be understood on the basis of the Hertz-Mindlin contact theory. However, such nonlinear friction theory does not adequately account for the behavior of energy dissipation observed in their experiments essentially independent of wave (strain) amplitude, a feature suggesting a linear mechanism for attenuation. This experimental observation raises an important question: Is it possible to reconcile a nonlinear frictional dynamics and a linear mechanism of dissipation at the grain contact level, particularly in the presence of adsorbed films?

In this Letter, we address this question by presenting new experimental results of elastic wave dissipation Q^{-1} obtained in dry and weakly wet granular materials by using codalike multiply scattered elastic waves [12]. Particular attention will be focused on the influence of surface properties of grains by adding a small amount of liquid or with adsorbed films. Compared to low-frequency waves, the high-frequency scattered waves have the advantage of separating the intrinsic attenuation from the scattering attenuation and infer the material properties on smaller length scales of the order of wavelength [13–15].

Experiments.—Our granular media are composed of slightly polydisperse glass beads of diameter $2R = 0.6$–0.8 mm manufactured for use as an abrasive (from Centraver). *Clean* granular samples are achieved by successively rinsing glass beads with acetone, ethanol, sulfochromic acid, and distilled water; the beads are then dried in a furnace. To study the effects of surface properties, small amounts of wetting liquid (volume fraction $\varphi_{\mathrm{liq}} = 0.05\%$) are added to glass beads. Here we used different silicon oils as wetting liquids to minimize the effects of evaporation. The kinematic viscosities of silicon oils (Rhodorsil 47V) ν_{liq} vary from 5 to 100 centistokes (cSt) due to their composition, possessing nevertheless the same density ρ_{liq} and tension surface γ_{liq}. The glass beads are mixed vigorously during tens of minutes to evenly distribute the liquid among the grains. Our glass beads are poured into a duralumin cylinder of diameter 30 mm and height 11 mm, layer by layer followed by successive compactions. A normal stress P_0 ranging from 0.1 to 1 MPa is then applied to the granular sample. Before each measurement, one cycle of loading and unloading is performed to the granular packing in order to consolidate the sample and minimize its hysteretic behavior. The solid volume fraction is about $\varphi_{\mathrm{sol}} \approx 63\%$ and 61% for dry and wet granular packings, respectively.

A longitudinal source transducer of diameter 30 mm and a small detecting transducer of diameter 2 mm are then placed on the axis of the cylindrical cell of diameter 30 mm in contact with glass beads. Fifteen-cycle tone burst excitations centered at $f\ (= \omega/2\pi) = 500$ kHz are applied

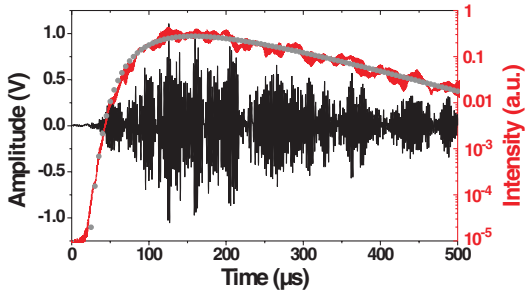

FIG. 1 (color online). Specklelike scattered ultrasound through a dry granular packing (black curve) and the transmitted intensity I averaged over 50 configurations [red (gray) curve]. Gray circles refer to the fit.

to the calibrated source transducer and the granular sample is subjected to 1 min of sound vibration before each acoustic measurement to ensure a better reversibility. Figure 1 illustrates a typical ultrasound transmission $s(t)$ through a dry glass bead packing under $P_0 = 0.62$ MPa and excited by the source displacement $u \approx 15$ nm, corresponding to an acoustic strain $\varepsilon(= \omega u/c_0) \approx 4.5 \times 10^{-5}$ with the compressional wave velocity $c_0 \approx 980$ m/s in the granular medium. The transmitted intensity of multiply scattered ultrasounds dominated by shear waves due to mode conversion [12], $I(t)(\sim\langle s(t)^2\rangle)$, is obtained by ensemble averaging over 50 independent configurations. By fitting the intensity profile with a diffusion model, we extract the internal dissipation Q^{-1} (the ratio of energy loss per cycle to peak energy stored) via the quality factor Q ($\approx 295 \pm 5$), and the diffusion constant D ($\approx 0.095 \pm 0.005$ m^2/s) [12].

We first investigate the effect of strain amplitude on the internal dissipation. Figure 2 represents Q^{-1} measured as a function of the source amplitude in dry and wet glass bead packings, respectively. In dry granular samples, cleaned or not, Q^{-1} increases linearly with strain indicating a nonlinear regime of dissipation, and tends to a finite constant at

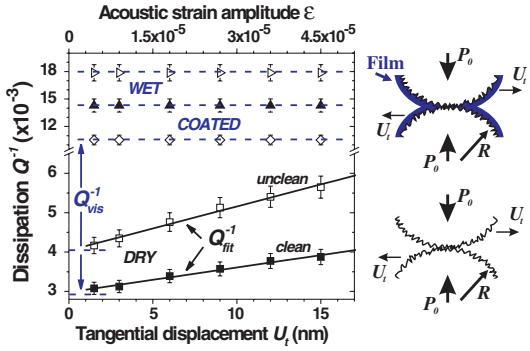

FIG. 2 (color online). Q^{-1} versus strain amplitude ε for different granular packings under $P_0 = 0.62$ MPa: clean (■), unclean (□), silanized (◇), and wet glass beads by silicon oil 47V10 (▲) and 47V100 (▷). Dashed lines refer to viscous behaviors $Q_{\rm vis}^{-1}$ (see the text), and solid lilnes to the fits Insets: Schemata of grain contacts between dry and coated beads by solid or liquid film.

small strain. Moreover, it is found that cleaning the glass beads permits one to reduce the dissipation, presumably due to the removal of dust layers from the grain surfaces.

In order to explore the effects of adsorbed film on grain surface, we conduct measurements of the Q^{-1} in a packing of glass beads coated by a hydrophobic film (silanized layer) of about 100 nm in thickness. The presence of the adsorbed solid film significantly increases the dissipation by a factor of about 3 as shown in Fig. 2, and results in a Q^{-1} nearly independent of strain amplitude over our experimental range. Similar behavior of dissipation is also found in the wet bead packing by silicon oils (Fig. 2). The wetting liquids form an adsorbed film of the order of the asperities (~100 nm) [16]. Measured Q^{-1} increases by a factor of 5–10, and the higher the liquid viscosity, the larger the dissipation Q^{-1}. In addition, Q^{-1} measured is again found to be independent of the strain amplitude, pointing to a dominant linear mechanism of dissipation. These results are different from those observed in dry media.

We next examine the effect of the applied effective stress P_0. In Fig. 3, we show Q^{-1} measurements versus the applied stress in dry and wet granular packings, respectively. As illustrated in Fig. 3(a), Q^{-1} measured for various strain amplitudes decreases clearly with increasing confining stress in dry media, a feature found in many earth materials [2,11]. Figure 3(b) shows a series of Q^{-1} measurements conducted in wet bead packings by adding silicon oil of different viscosity. Surprisingly, Q^{-1} is found to *increase* with increasing confining stress, displaying inverse dependence on P_0 compared to that observed in dry granular media.

Analysis and discussion.—To account for the experimental observation of elastic wave dissipation, we compare the measured Q^{-1} with some of the predictions by the contact theory. As a first approach to the problem, the model for the glass bead packing may be taken as a homogeneous random contact network of uniform spheres $2R$. The bulk elastic and dissipative responses of the granular system are thus directly related to the dynamic properties of the individual contact (cf. insets of Fig. 2). When such a contact is submitted to a shear force $F_t \leq \mu F_n$ with

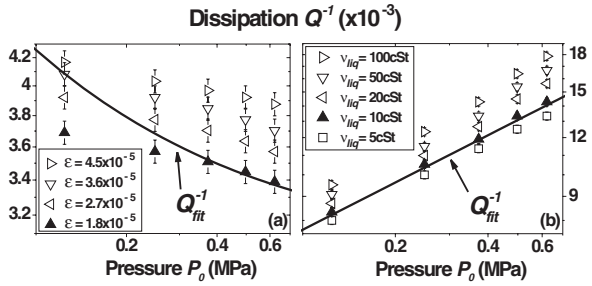

FIG. 3. Q^{-1} versus applied stress P_0 in different granular packings (in log-log scale). (a) Dry (clean) beads for different strain amplitudes ε. (b) Weakly wet beads with different kinematic viscosity $\nu_{\rm liq}$ of silicon oils. Solid lines refer to the fits (see the text).

F_n the normal load and μ the friction coefficient, Mindlin has shown that the interfacial normal and shear stiffness, k_n and k_t, are comparable. For the oscillating amplitudes of normal and shear displacement at the contact surfaces, U_n and U_t, the peak strain energy stored is $W = \frac{1}{2}k_n U_n^2 + \frac{1}{2}k_t U_t^2$. The energy dissipation per cycle ΔW at each contact should then be evaluated in order to determine $Q^{-1} = \Delta W/(2\pi W)$.

In the dry bead packing, we examine the prediction by the Mindlin theory, the most widely used model describing the friction between two *smooth* spheres. At low oscillating amplitude ΔW_{fric} is proportional to F_t^3 or U_t^3 [17] and Q_{fric}^{-1} is written as

$$Q_{\text{fric}}^{-1} \approx \left[\frac{4G}{9\pi^2(2-\nu)R}\left(\frac{3\pi}{4E^*}\right)^{1/3}\right]\mu^{-1}P_0^{-2/3}U_t, \quad (1)$$

where ν, E^*, and G are the Poisson ratio, bulk, and shear modulus of the glass material, respectively, and $P_0 = F_0/(\pi R^2)$. The most prominent feature of Eq. (1) is the amplitude dependence of Q_{fric}^{-1}, characteristic of a nonlinear frictional attenuation. This theory provides a simple interpretation of the increasing of Q^{-1} with the strain amplitude. Note that such nonlinear *hysteretic* attenuation is also found in a large class of earth materials [18]. However, the frictional dissipation at small strains either disappears or becomes masked by amplitude-independent (linear) dissipation mechanism as observed in Fig. 2. Similarly, a linear dissipation regime was evidenced for small-strain oscillation of a steel sphere in contact with a flat glass surface by Johnson [17], and between two identical rough solids by Bureau *et al.* [19]. The loss mechanism is ascribed to the linear *viscoelastic* dissipation within the bodies of contacting asperities.

As a result, the interfacial dissipation is described by the interplay of two mechanisms:

$$Q^{-1} = Q_{\text{fric}}^{-1} + Q_{\text{vis}}^{-1}. \quad (2)$$

Using the material constants of glass, $G = 28$ GPa, $E^* = 37$ GPa, $\nu = 0.25$, one can extract by fitting Eq. (2) both the Q_{vis}^{-1} and the friction coefficient μ. For $P_0 = 0.62$ MPa, one obtains by fit $Q_{\text{vis}}^{-1} \approx 4 \times 10^{-3}$ and $\mu \approx 0.98$ for *unclean* bead packing, and $Q_{\text{vis}}^{-1} \approx 3 \times 10^{-3}$ and $\mu \approx 1.75$ for *clean* bead packing, respectively. Note that the deduced Q_{vis}^{-1} is much larger than that of the bulk glass Q_{glass}^{-1} ($\sim 10^{-6}$) [20], pointing thus to the interfacial dissipation which we speculate is due to the viscoelastic loss within a contacting layer different from the bulk of the glass bead. Moreover, as expected, the friction coefficient of unclean beads is smaller than that of clean beads, probably due to the presence of the dust film on glass bead surfaces which form effective lubricants and enhance sliding dissipation [11]. However, the absolute values of the friction coefficient deduced are unusually high ($\mu_{\text{glass}} \approx 0.3$). As revealed in a recent work [19], the Mindlin model fails to describe quantitatively the dissipative response on rough interfaces, because the use of local friction coefficient is legitimate only when the width L of the microslip zone is much larger than hundreds of nm. However, this condition is *not* satisfied by the small-amplitude shear oscillating, e.g., $\varepsilon \approx 10^{-5}$, which produce merely a L of about 300 nm.

On the other hand, Eq. (1) predicts that Q_{fric}^{-1} decreases with increasing confining stress as $P_0^{-2/3}$. Such dependence occurs for two reasons. First, increasing P_0 stiffens the frame moduli, thereby increasing strain energy as $W \sim P_0^{1/3}$. Second, ΔW_{fric} decreases with increasing P_0 as $P_0^{-1/3}$. Figure 3(a) shows the Q_{fit}^{-1} estimated from Eqs. (1) and (2) with the fit parameter μ for clean beads and acoustic displacement $U_t = 6$ nm (or strain $\varepsilon = 1.8 \times 10^{-5}$) which agrees qualitatively well with the experiments at high pressure, but not at low pressure. This discrepancy may arise from various factors mentioned above, but also from the fact that the distribution of contact force network is particularly inhomogeneous under low pressure, thus deviated significantly from the mean-field approximation assumed in this work [6].

In the granular packing composed of *coated* beads either with a viscoelastic silanized layer or with wetting liquid films, the linear viscous dissipation largely dominates the frictional one, as shown in Fig. 2. The silanized glass exhibits lower friction coefficient [19], but the nonlinear frictional dissipation is masked even at the largest strain amplitude used here $\varepsilon = 4.5 \times 10^{-5}$, by the linear mechanism of dissipation $Q_{\text{vis}}^{-1} \approx 10^{-2}$, likely related to the viscoelastic loss within the body of solid films.

We now focus on the energy loss in the wet granular packing by adding liquids—a major result of this work. Halsey and Levine [21] proposed a qualitative picture for describing how the wetting liquid occupies the microscopically rough surfaces between two spheres as a function of the added-liquid volume. For small amounts of wetting liquids added to the granular packing, i.e., liquid volume fraction $\varphi_{\text{liq}} \sim 0.05\%$ as that used in our work, the wetting liquid remains *trapped* in tiny menisci at the surface asperities and is capable of resisting the contact between grains. The thickness of adsorbed liquid film at the grain surfaces is roughly about the height of asperity (inset of Fig. 2), confirmed by the microscopic observations [16,22].

We then propose the following picture. When high-frequency elastic waves pass, the contact surfaces begin to slide past each other due to the important reduction of shear strength by boundary-layer and hydrodynamic lubrications of thin liquid films, left in the gap between contact surfaces. These liquid films contribute an additional linear viscous dissipation Q_{liq}^{-1} in Eq. (2) which appears predominant in the wet bead packings (Fig. 2). To characterize such a loss, we calculate the dissipated energy ΔW_{liq} by shearing a thin liquid film of thickness δ_{liq} and area Σ_{liq}, confined at the contact surface: $\Delta W_{\text{liq}} \approx (2\pi)\rho_{\text{liq}}\nu_{\text{liq}}(\Sigma_{\text{liq}}/\delta_{\text{liq}})\omega U_t^2$. As the shear stiffness of a sliding contact vanishes ($k_t \sim 0$), the strain energy stored at the contact is $W = \frac{1}{2}k_n U_n^2 \approx \frac{1}{4}k_n U_0^2$. Approximating Σ_{liq} by

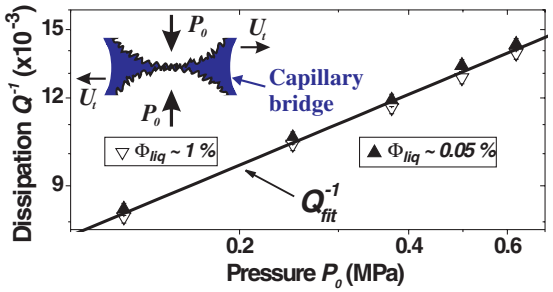

FIG. 4 (color online). Q^{-1} versus applied stress P_0 for granular packings wet by silicon oil with two different liquid volume fractions (in log-log scale): $\varphi_{\mathrm{liq}} = 0.05\%$ (▲) and $\varphi_{\mathrm{liq}} = 1\%$ (▽). Solid line refers to the fit (see the text). Inset: Schema of liquid filling between grains.

the Hertz contact area ($\Sigma_{\mathrm{liq}} \sim P_0^{1/3}$) yields

$$Q_{\mathrm{liq}}^{-1} \approx \left[\frac{(2-\nu)\pi R}{2G\rho_{\mathrm{liq}}}\left(\frac{4E^*}{3\pi}\right)^{1/3}\right]\omega\nu_{\mathrm{liq}}\delta_{\mathrm{liq}}^{-1}P_0^{1/3}, \quad (3)$$

which is independent of strain amplitude. On the other hand, Eq. (3) predicts that Q_{liq}^{-1} increases with increasing confining stress as $P_0^{1/3}$, a scaling behavior *opposite* to that expected by the frictional loss [Eq. (1)]. This feature is due to the dissipated energy ΔW_{liq}, which increases with confining stress as $P_0^{2/3}$.

In addition, we can estimate the thickness δ_{liq} from the measurements via Eq. (3), which is about 10–200 nm for $\nu_{\mathrm{liq}} = 5$–100 cSt. These values are reasonably in agreement with the roughness of the abrasive glass beads [16]. To ensure that the viscous dissipation is primarily due to the adsorbed liquid films *sheared* at the contact area and not due to the bulk liquid flow, we have measured Q^{-1} in a wet granular sample by adding liquids up to $\varphi_{\mathrm{liq}} \sim 1\%$. For such liquid filling, capillary menisci are formed at the grain contact as shown in the inset of Fig. 4 [21]. However, no difference of Q^{-1} is measured between these two liquid fillings, confirming that the viscous dissipation occurs predominantly in the adsorbed film.

In summary, our experiments reveal the crucial role of the adsorbed films, liquid or solid, on the dissipation of elastic waves in dry and weakly wet granular media. We have identified two distinct sources of dissipation from the adsorbed films (impurity layers) in dry media: a boundary lubrication effect which enhances the nonlinear frictional loss, and a viscoelastic effect which contributes to linear dissipation. For submicron-thick adsorbed films investigated in this work, liquid or solid, the linear viscous dissipation is the dominant mechanism in the contribution to the observed dissipation. Clearly, further study is needed to understand the interplay of the linear and nonlinear regimes of dissipation as a function of the structure and interfacial properties of the adsorbed film [23].

We believe that our work points to the considerable interest in scattered-wave probing as a tool for studying the nature of grain surfaces in granular mechanics, such as singing sands [24], and the effects of humidity-induced capillary condensation in wet granular materials, including room-dry rocks.

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