Nonlinear Elasticity of Granular Materials

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The elastic properties of granular materials can be enormously nonlinear as compared with the properties of non-porous materials. Experiments on isotropic compression of a granular assembly of spheres show that the shear, μ, and bulk, k, moduli vary with the confining pressure faster than the 1/3 power law predicted by Hertz-Mindlin elasticity theory. Moreover, the ratio between the experimental bulk and shear moduli is found to be constant but with a value larger than the theoretical prediction. Numerical simulations resolve the question as to whether the problem lies with the treatment of the grain-grain contact or with the elastic framework. We find that the problem lies principally with the later: the *affine* assumption (which underlies the elastic formulation) is found to be valid for k but to breakdown seriously for μ. This explains why the experimental and numerical values of $\mu(p)$ are much smaller than the elastic predictions. In this paper we review recent progress on the understanding of this problem based on microscopic simulations, elasticity theory, and more innovatives ideas such as jamming, fragility and thermodynamics of granular materials.

I. INTRODUCTION

The study of elastic properties of granular materials [1,2] has been treated by many researchers since the pioneering work of Mindlin in the 50's [3–12]. However, a basic understanding of the physics of nonlinear granular elasticity is currently lacking [1].

In a typical experiment, a set of glass beads is confined at a hydrostatic pressure, p, and the compressional sound speed, v_p, and the shear sound speed, v_s, are measured as a function of pressure (see for instance Domenico [6], Yin [7], and [5,9,12]). The P-wave and S-wave speeds are related to the elastic constants of the material: $v_p = \sqrt{(k + 4/3\mu)/\rho}$ and $v_s = \sqrt{\mu/\rho}$, where k and μ are the bulk and shear moduli of the system and ρ is the density of the system.

Important insight into this problem comes first from the Hertzian contact theory to model the intergrain forces [13]. In this case, nonlinearity arises from the increase with the external pressure of the contact area between two spherical grains. Other nonlinearities arise due to the fact that the number of contacts per grain increases as the confining pressure increases.

Conventional theories to describe this problem are based on the theory of elasticity of continuum media developed for non-porous materials [14]. Inherent to an elastic formulation is a uniform strain at all scales and that the displacement field of the grains is affine with the macroscopic deformation. A common approximation used in this theory is to consider the disordered medium as an effective medium that exerts a mean-field force on a single representative grain. This approximation is usually referred to as Effective Medium Theory (EMT) [3,4].

It is found experimentally that the shear and bulk moduli of an assembly of spherical grains vary with the confining pressure, p, faster than the power law predicted by conventional elasticity theory (EMT) based on Hertz-Mindlin contact mechanics [3,4,12]. EMT predicts that both moduli vary as $k_e \sim \mu_e \sim p^{1/3}$, and that the ratio k_e/μ_e is a constant independent of pressure, independent of coordination number, and dependent only on the Poisson's ratio of the individual grains. [2] The value of k/μ found experimentally, though constant, is larger than the EMT prediction.

The origin of the above discrepancies has not been clear. These discrepancies between theory and experiment could be due to the breakdown of the Hertz-Mindlin force law at each grain contact as or they could be associated with the breakdown of the continuum theory of elasticity. de Gennes [10] proposed that a thin shell of oxide layer would give a faster grow with pressure of the elastic moduli of the system, which may explain the behavior for metallic beads. Goddard [5] proposed that sharp angularities of the grains (for instance sand grains) may modify the contact law force between grains, giving rise to a different pressure dependence than the predicted by Hertzian theory which is valid for spherical grains. Other authors [5,11] have suggested that the increasing number of contacts with pressure may be the

[1]Here, nonlinear effects refer to the fact that the elastic moduli— calculated using small-amplitude strains— depends on the external pressure, and they do not refer to the nonlinear mechanical response to large strain perturbations.

[2]The subscript denotes that these values are calculated considering granular materials as purely elastic solids.

reason for the discrepancies in the pressure dependence of the moduli. Jenkins *et al.* [11] measured the elastic moduli using numerical simulations and concluded that EMT does not describe correctly the shear modulus but it describes fairly well the bulk modulus. Other experimental work done by Liu and Nagel [8] and Jia *et al.* [9] concentrated on the role played by force chains in sound propagation. Interesting approaches based on original ideas by Nesterenko for propagating waves in one dimensional elastic chains has been also applied to wave propagation in granular media [15,16].

There is a clear need for a microscopic study in order to explore the above discrepancies. In collaboration with D. Johnson and L. Schwartz from Schlumberger and N. Gland from Ecole Normale Supérieure we have performed molecular dynamics (MD) simulations for a system of unconsolidated spherical glass beads. We were able to calculate the elastic properties of a disordered array of Hertz-Mindlin spherical grains [12].

We found agreement between our simulations and existing experimental data, thus confirming the validity of the Hertz-Mindlin contact theory to glass bead aggregates. Our study quantifies two problems with the conventional elastic continuum approach: *(i)* If the calculated increase of the average coordination number with increasing pressure is taken into account, the modified EMT gives an accurate description of the bulk modulus, $k(p)$, but it gives only the correct trend of the pressure dependence of the shear modulus, $\mu(p)$. *(ii)* Most importantly, we showed that the affine assumption is approximately valid for the bulk modulus and seriously in error for the shear modulus; this fact is why the EMT prediction of k_e/μ_e differs significantly from the experimental value. We conclude that in order to develop a better understanding of the problem, one must abandon the purely elastic framework and consider granular matter as a full viscoelastic body. Viscoelastic effects can account for the discrepancy in the shear modulus in comparison with the elastic prediction: the corrections increase dramatically in the case of loose materials or for weak intergrain shear forces. The complex relaxation mechanism is related to the structural disorder of the packing structure, such as heterogeneities in the contact force network or "force chains" (stress paths of grains carrying most of the forces in the material [17–19]), and the nonaffine motion of grains.

Here we briefly describe our efforts to understand the nonlinear elasticity of granular materials. We will review also recent approaches to this problem based on perturbative studies of the mean-field approximation and more innovative approaches based on thermodynamics of granular matter, as well as concepts such as jamming and fragility.

II. THEORIES OF GRANULAR ELASTICITY

In the microscopic model two spherical grains with radii R_1 and R_2 interact via a normal Hertz force, $F_n = \frac{2}{3}\ k_n R^{1/2}\xi^{3/2}$, and a transverse Mindlin force, $\Delta F_t = k_t(R\xi)^{1/2}\Delta s$ [13]. Here $R = 2R_1R_2/(R_1 + R_2)$, the normal overlap is $\xi = (1/2)[(R_1+R_2)-|\vec{x}_1-\vec{x}_2|] > 0$, and $\vec{x}_1, \vec{x}_2$ are the positions of the grain centers. The normal force acts only in compression, $F_n = 0$ when $\xi < 0$. The variable s is defined such that the relative shear displacement between the two grain centers is $2s$. The prefactors $k_n = 4\mu_g/(1-\nu_g)$ and $k_t = 8\mu_g/(2-\nu_g)$ are defined in terms of the shear modulus μ_g and the Poisson's ratio ν_g of the material from which the grains are made (typically $\mu_g = 29$ GPa and $\nu_g = 0.2$, for spherical glass beads). Internal dissipation is included in two ways: (1) via a viscous damping term proportional to the relative normal and tangential velocities, and (2) via sliding friction: when F_t exceeds the Coulomb threshold, $\mu_f F_n$, the grains slide and $F_t = \mu_f F_n$, where μ_f is the kinematic friction coefficient between the spheres (typically $\mu_f = 0.3$). We usually assume a distribution of grain radii in which $R_1 = 0.105$ mm for half the grains and $R_2 = 0.075$ mm for the other half.

The basic idea of elastic theories relevant to our study is that the macroscopic work done in deforming the system is set equal to the sum of the work done of each grain-grain contact and that the latter is replaced by a suitable average [3,4].

In the case of an isotropic deformable solid, the energy of the assembly as a function of the strain ϵ_{ij} is [14] $U = \mu_e(\epsilon_{ij} - \frac{1}{3}\delta_{ij}\epsilon_{ll})^2 + k_e\epsilon_{ij}^2$, and the work done by the imposed strain is calculated in terms of the intergrain forces and displacements. There are two assumptions inherent to the elastic EMT:

(1) All the spheres are statistically the same, and it is assumed that there is an isotropic distribution of contacts around a given sphere. (2) An affine approximation is used, i.e., the spheres at position X_j are moved a distance δu_i in a time interval δt according to the macroscopic strain rate $\dot{\epsilon}_{ij}$ by

$$\delta u_i = \dot{\epsilon}_{ij} X_j \delta t. \tag{1}$$

The grains are always at equilibrium due to the assumption of isotropic distribution of contacts and further relaxation is not required. This sort of mean field theory is analogous to a simple average of the non-linear spring constants.

The EMT predictions for the bulk and shear modulus are:

$$k_e(p) = \frac{k_n}{12\pi}\ (\phi Z)^{2/3}\ \left(\frac{6\pi p}{k_n}\right)^{1/3}, \tag{2a}$$

$$\mu_e(p) = \frac{k_n + \frac{3}{2}k_t}{20\pi}\ (\phi Z)^{2/3}\ \left(\frac{6\pi p}{k_n}\right)^{1/3}, \tag{2b}$$

where ϕ is the volume fraction and Z the average coordination number of the spheres.

As discussed above, the $p^{1/3}$ dependence of the moduli is not corroborated by experiments. Another way of seeing the breakdown of the elastic theory is to focus on the ratio k/μ. According to Eqs. (2), $k_e/\mu_e = 5/3(2-\nu_g)/(5-4\nu_g)$, independent of pressure, a value which depends only on the Poisson's ratio of the bead material. The experiments give $k/\mu \approx 1.1 - 1.3$. Our simulations give $k/\mu \approx 1.05 \pm 0.1$ [12] in good agreement with experiments. EMT predicts $k_e/\mu_e = 0.71$, if we take $\nu_g = 0.2$ for the Poisson's ratio of glass. [The EMT prediction is quite insensitive to variations of ν_g; $k_e/\mu_e = 0.71 \pm 0.04$ for $\nu_g = 0.2 \pm 0.1$.] Conversely, a value $\nu_g \simeq 1.2$ would be needed in order to fit the experimental k/μ clearly violating the upper thermodynamic limit on $\nu_g \leq 1/2$ [14].

III. NUMERICAL RESULTS

We perform MD simulations of systems of the order of 10000 particles [12]. A model used in current granular simulations is the Discrete Element Method developed by Cundall and Strack [20,21] for a system of soft spheres [13]. Our calculations begin with a numerical protocol designed to mimic the experimental procedure used to prepare dense packed granular materials. Generating a mechanically stable packing is not a trivial task. The simulations begin with a gas of spherical particles located at random positions in a periodically repeated cubic cell of side L. At the outset, a series of strain-controlled isotropic compressions and expansions are applied until a volume fraction slightly below the critical density is reached. The system is then compressed and extended slowly until a specified value of the pressure and volume fraction— above the random close packing (RCP) fraction— is achieved at static equilibrium.

Consider, now, the calculation of the elastic moduli of the system as function of pressure. Beginning with the equilibrium state describe above, we apply a small perturbation to the system and measure the resulting response. The shear modulus is calculated in two ways, from a pure shear test, $\mu = (1/2)\Delta\sigma_{12}/\Delta\epsilon_{12}$, and also from a biaxial test, $\mu = (\Delta\sigma_{22} - \Delta\sigma_{11})/2(\Delta\epsilon_{22} - \Delta\epsilon_{11})$. The bulk modulus is obtain from a uniaxial compression test, $k + 4/3\mu = \Delta\sigma_{11}/\Delta\epsilon_{11}$. Here the stress, σ_{ij}, is determined from the measured forces on the grains, and the strain, ϵ_{ij}, is determined from the imposed dimensions of the unit cell.

In Fig. 1 our calculated values of the elastic moduli as a function of pressure are compared with EMT and with experimental data. From Fig. 1 we see that our experimental and numerical results are in reasonably good agreement. Also shown are measured data from Domenico [6] and Yin [7]. Clearly, the experimental data are somewhat scattered. This scatter reflects the difficulty of the measurements, especially at the lowest pressures where there is significant signal loss. Nevertheless, our calculated results pass through the collection of available data. Also shown in Fig. 1 are the EMT predictions (2). The EMT curves are obtained using the same parameters as in the simulations; we also set $Z = 6$ and $\phi = 0.64$, independent of pressure. At low pressures we see that k is well described by EMT. At larger pressures, however, the experimental and numerical values of k grow faster than the $p^{1/3}$ law predicted by EMT. The situation with the shear modulus is even less satisfactory. EMT overestimates $\mu(p)$ at low pressures but, again, underestimates the increase in $\mu(p)$ with pressure.

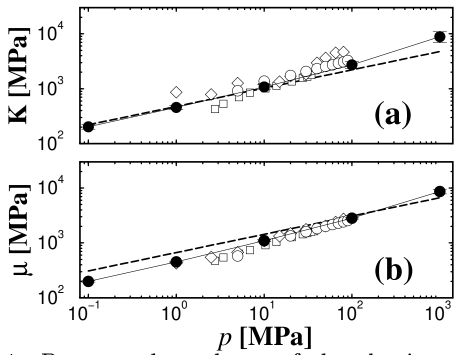

FIG. 1. Pressure dependence of the elastic moduli from MD (•), experiments [Domenico (□) [6], Yin (◇) [7], and our experiments (◯)], and theory (dashed line): (a) bulk modulus, and (b) shear modulus.

To understand why μ is over predicted by EMT we must examine the role of transverse forces and rotations in the relaxation of the grains. [These effects do not play any role in the calculation of the bulk modulus.] Suppose we re-define the transverse force by introducing a multiplicative coefficient α, viz: $\Delta f_t = \alpha k_t (R\xi)^{1/2}\Delta s$; with $\alpha = 1$ we recover our previous results. To quantify the role of the transverse force on the elastic moduli, we calculate $k(\alpha)$ and $\mu(\alpha)$ at a given pressure $p = 100$ KPa [Fig. 2a]. This pressure is low enough that the changing number of contacts is not an issue. Surprisingly, the shear modulus becomes negligible small as $\alpha \to 0$. As expected, k is independent of the strength of the transverse force. To compare with the theory we also plot the prediction of the EMT, Eq. (2) in which k_t is rescaled by αk_t. We see that the EMT fails in taking into account the vanishing of the shear modulus as $\alpha \to 0$. However it accurately predicts the value of the bulk modulus, which is independent of α.

Figure 2b shows the typical viscoelastic response of the granular medium. In the MD calculation of the shear modulus an affine perturbation is first applied to the system. The shear stress increases (from A to B in Fig. 2b) and the grains are far from equilibrium since the system is disordered. The grains then relax towards equilibrium (from B to C), and we measure the resulting change in stress from which the modulus is calculated. To better understand the approximations involved in the EMT, suppose we repeat the MD calculations taking into account only the affine motion of the grains and ignoring the subsequent relaxation. The resulting values of the moduli are plotted in Fig. 2a as open symbols and we see that the moduli calculated this way are very close to the EMT predictions. Thus, the difference between the MD and EMT results for the shear modulus lies in the non-affine relaxation of the grains; this difference being largest when there is no transverse force. By contrast, grain relaxation after an applied *isotropic* affine perturbation is not particularly significant and the EMT predictions for the bulk modulus are quite accurate.

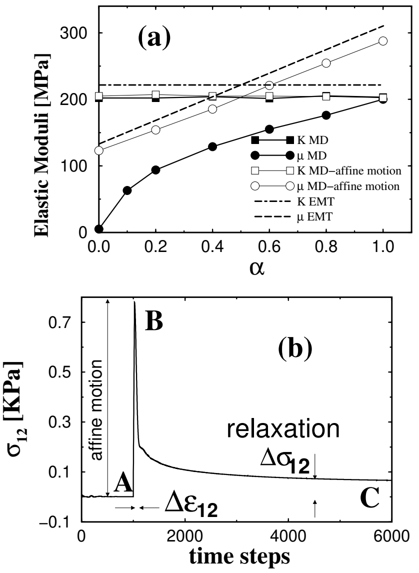

FIG. 2. (a) $k(\alpha)$, and $\mu(\alpha)$ versus α for a fixed $p = 100$ KPa. (b) Relaxation of the shear stress (B→C) after an affine motion (A→B) in the calculation of the shear modulus.

The surprisingly small values of μ found as $\alpha \to 0$ can be understood as a melting of the system that occurs when the system is close to the RCP fraction. This fluid like behavior (when $k_t = 0$) is closely related to the melting transition seen in compressed emulsions [22] and foams [23]. At the RCP fraction the system behaves like a fluid with no resistance to shear, and might indicate that the system is in a fragile state.

Our results can be analyzed in terms of jamming [24,25] theory. It has been proposed that jamming— and in some particular cases fragility [26]— are universal features of many complex systems with a common slow relaxation dynamics such as glasses, foams, and granular matter [24,25]. We notice that granular systems can jam at RCP [27] but also at other states of lower density known as random loose packing (RLP) states [28]. It is near the jamming transition that we expect to find the most interesting viscoelastic properties.

In the limit of weak compression, the stress vanishes continuously as $\sigma \sim (\phi - \phi_c)^\beta$, where ϕ_c is the critical density of the jamming transition [19,29]. Jamming can occur at RLP or RCP according to the existence or not of transverse forces between the grains, respectively. At the jamming transition, the coordination number approaches a minimal value Z_c (=4 for friction and 6 for frictionless grains) also as a power law. Our result $Z_c = 6$ agrees with experimental analysis of Bernal packings for close contacts between spheres fixed by means of wax, and our own analysis of the Finney packings [27] using the actual sphere center coordinates of 7934 steel balls. However, no similar experimental study exists for RLP which could be able to confirm $Z_c = 4$. A critical slowing down— the time to equilibrate the system increases near ϕ_c— and the emergence of shear rigidity is also found at criticality.

Concomitant with the approach to the jamming transition we find a change in the probability distribution and in the degree of spatial correlation of the interparticle forces: localized force chains with an exponential probability distribution— as observed in recent studies [18]— are found at low confining stress giving way to a considerably less localized and homogeneous arrangement of the forces with a Gaussian probability distribution at higher stress away from the critical density [19].

IV. THERMODYNAMICS FORMULATION OF GRANULAR MATERIALS

In equilibrium statistical mechanics of fluids microscopic information on the fluctuations of the particle motion equilibrated at temperature T is related to macroscopic dissipative variables, such as the viscosity, via the fluctuation-dissipation theorem (FDT), that in one of its forms — Stokes-Einstein relation— relates the random diffusion of thermally fluctuating positions of particles, and the response function to an external force [30]. Even though the diffusivity of the tracer D and mobility χ are strongly dependent on size and shape of the tracer, they turn out to be always related by the Einstein relation $D/\chi = T$, where T is the temperature of the liquid.

One may wonder if this approach could be applied to an out-of-equilibrium system such as granular matter. If so, we could have a theoretical framework for understanding macroscopic quantities such as complex shear modulus from detailed microscopic variables.

At first sight, investigations of a thermodynamics relation such as FDT for granular materials of size larger than 1 μm might seem to be condemned to fail: granular materials are athermal systems characterized by the fact that energy is supplied by external driving (via shear or tapping) and dissipated by inelastic interactions and slippage between the grains— a very different "heat exchange mechanism" from that of a thermal bath in a molecular or colloidal system. However, recent theoretical and numerical work has yielded mounting evidence of an underlying thermodynamics valid for dense granular media. This train of ideas started more than a decade ago by S. Edwards and collaborators [31]. Edwards proposed a thermodynamics and statistical ensemble valid for dense slowly moving granular materials and glasses, and through it, thermodynamic notions of entropy and temperature.

The thermodynamic construction of Edwards is based on two postulates [31]: (1) While in the Gibbs construction for equilibrium statistical mechanics one assumes that the physical quantities are obtained as average over all possible configurations at a given energy, Edwards ensemble consists of only the blocked configurations (static or jammed) at the appropriate energy and volume. (2) The strong 'ergodic' hypothesis is that all blocked configurations of given volume and energy can be taken to have equal statistical weights. This formulation leads to an entropy $S_{\rm Edw}(E,V)$ as the logarithm of the number of blocked configurations, and the corresponding temperature

$$T_{\rm Edw}^{-1} = \frac{\partial S_{\rm Edw}}{\partial E}, \tag{3}$$

and compactivity $X_{\rm Edw}^{-1} = \partial S_{\rm Edw}/\partial V$.

Recent theoretical support to this formulation came from glass theory [32,33]: theoretical and numerical work on schematic models has yielded evidence supporting this thermodynamic picture— and this has spurred a renewed interest in Edwards' thermodynamics. Indeed, recent analytic schemes for out of equilibrium glassy dynamics have suggested the existence of a FDT thermodynamical temperature (unrelated to the room temperature) governing the slow components of fluctuations and responses of all observables in the glassy state [34,32]. Furthermore, this fluctuation-dissipation temperature has been shown to be directly related to Edwards' ideas for a statistical ensemble for granular matter [31]— within schematic models [33] and in MD models of granular matter [35].

We have recently performed in collaboration with J. Kurchan, with a realistic numerical model, a diffusion-mobility numerical experiment [35]. Our results support the existence of a notion of temperature for long-scale displacements (structural rearrangements) independently of the viscous friction dissipation between grains or even the presence sliding friction (Coulomb law).

We performed MD simulations for a binary system of large and small spheres in a periodic 3D cell. We applied a gentle simple shear flow at constant volume and measure the spontaneous fluctuations $\langle \Delta x^2 \rangle$ and force-induced displacements $\langle \Delta x \rangle / f$, where f is an small external force, for two types of tracers with different sizes. Our data was consistent with an extended Einstein relation:

$$\langle \Delta x^2 \rangle = 2\, T_{\rm FDT} \frac{\langle \Delta x \rangle}{f}, \tag{4}$$

valid for both particles *with the same* $T_{\rm FDT}$ for widely separated time scales. This suggested that $T_{\rm FDT}$ can be considered to be the temperature of the slow modes.

We then performed an explicit computation to show that the temperature arising from Edwards' thermodynamic description and the dynamic one measured from Einstein's relation coincide. This last step cannot be performed in the laboratory, so the numerical simulation provides the missing link between thermodynamic ideas and diffusion-mobility checks. In order to calculate $T_{\rm Edw}$ and compare with $T_{\rm FDT}$, we counted the number of blocked configuration at a given energy and volume. To do this in practice, we extended the 'auxiliary model' method of [33] to the case of deformable grains.

This work strongly supports the thermodynamic picture. A hypothesis being currently tested is whether the temperature arising from Edwards formalism may provide the link between the mechanical properties and the microstructural relaxation properties of granular materials. Based on our previous studies, we consider a generalized Stokes-Einstein relation to finite frequencies: $\tilde{G}_s = s\,\tilde{\eta}_s = T_{\rm FDT}/[\pi R s \langle \Delta\, \tilde{x}_s^2 \rangle]$. Here one extract the Laplace transform of the shear modulus $\tilde{G}_s$ of the viscoelastic material by measuring the power spectrum of the root-mean square displacement of a tracer (of size R) moving in a viscoelastic medium. The frequency-dependent GSER relates then the fluctuation of the tracer to the medium macroscopic linear viscoelasticity $G^*(w)$ obtained via analytic continuation $s = i\omega$.

This approach is motivated by studies of microrheology in complex fluids. In 1995 Mason and Weitz [36] developed a technique, called microrheology, using a GSER to extract the complex shear modulus from the equilibrium thermal fluctuations of a tracer moving in the viscoelastic material. In the case of athermal slowly driven granular materials, the thermodynamic Edwards temperature provides the link between the microscopic quantities and the shear modulus.

V. PERTURBATIVE EMT

Additionally we have carried out a perturbative approach in collaboration with J. Jenkins from Cornell University. Here a pair fluctuation analysis is employed where the relaxation of a pair of particles immerse in the effective medium is considered, instead of the response of a single particle as in the original EMT. In this way, effects of relaxation can be incorporated in the elastic theory. The theoretical corrections to the moduli are given in terms of two tensorial quantities such as $A_{ij} = \sum_{\rm all\ pairs} k'_n n_i n_j$ and $J_{ijk} = \sum_{\rm all\ pairs} k'_n n_i n_j n_k$, which depend on the packing structure (k'_n is a pressure-dependent stiffness). These tensors are obtained as a function of the pressure with our MD simulations, to study the corrections to EMT in the entire pressure range. Preliminary computational results indicate that corrections could become relevant in the case of frictionless grains.

VI. SUMMARY

Our MD simulations are in good agreement with the available experimental data on the pressure dependence of the elastic moduli of granular packings. They also serve to clarify the deficiencies of EMT. Grain relaxation after an infinitesimal affine strain transformation is an essential component of the shear (but not the bulk) modulus. This viscoelastic behavior is not taken into account in the EMT. In the limit $\alpha \to 0$ a packing of nearly rigid particles responds to an external isotropic load with an elastic deformation and a finite k. By contrast, such a system cannot support a shear load ($\mu \to 0$) without severe particle rearrangements. This may indicate a "fragile" state of the system [26] where inter particle forces are organized along "force chains" (stress paths carrying most of the forces in the system) oriented along the principal stress axes. Such fragile networks support, elastically, only perturbations compatible with the structure of force chains and deform plastically otherwise. Clearly, there is a need for an improved theory; recent work on thermodynamics and jamming of granular materials may provide an alternative formulation and allow to describe properly the response of granular materials to perturbations.

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