Hysteresis and Competition between Disorder and Crystallization in Sheared and Vibrated Granular Flow

Karen E. Daniels and Robert P. Behringer

Department of Physics and Center for Nonlinear and Complex Systems, Duke University, Durham, North Carolina 27708, USA

(Received 4 October 2004; published 29 April 2005)

Experiments on spherical particles in a 3D annular shear cell vibrated from below and sheared from above show a hysteretic freezing or melting transition. Under sufficient vibration a crystallized state is observed, which can be melted by sufficient shear. The critical line for this transition coincides with equal kinetic energies for vibration and shear. The force distribution is double peaked in the crystalline state and single peaked with an approximately exponential tail in the disordered state. Continuous relations between pressure and volume (with $dP/dV > 0$) exist for a continuum of partially and/or intermittently melted states over a range of parameters.

DOI: 10.1103/PhysRevLett.94.168001 PACS numbers: 45.70.-n, 64.60.Cn, 64.60.My, 64.70.-p

The relative stability and selection of crystallized and disordered states is fundamental to the study of condensed matter systems. For systems out of equilibrium, a thermodynamic picture under which temperature melts crystalline order need not be valid. Fluctuations provided by external driving, seemingly temperaturelike, may either order or disorder a system. In particular, the mechanisms by which colloidal suspensions shear-melt and shear-order [1,2] remain the subject of debate. Granular materials, which are athermal and have strongly dissipative interactions, provide a complementary means to investigate the transition between disorder and crystallization in nonequilibrium systems.

We experimentally examine a granular system exhibiting a novel phase transition involving competition between ordering and disorder through two different types of energy input. We use a variation on a Couette cell, a convenient setup for the study of sheared granular materials [3–5]. An annular region containing monodisperse spheres is vibrated from below and sheared from above, with the mean pressure and volume set from below by a spring. The shear and vibration provide competing effects leading to disorder or crystallization depending on their relative strength; the boundary between these two states occurs at equal kinetic energy input from these two driving mechanisms. This transition is hysteretic, similar to freezing-by-heating [6]. We also observe metastable states over a range of packing fractions and forces with the remarkable property that increased internal forces occur for less dense packings, in contrast to ordinary mechanical or thermodynamic compressibility. While previous studies have investigated vibrated [7] or continuously sheared [8] granular materials separately, the combination of the two leads to novel effects.

Figure 1 shows a schematic of the apparatus. Monodisperse polypropylene spheres fill the region between two concentric, stationary side walls, with a motor-driven rotating upper plate and a pistonlike bottom plate attached to an electromagnetic shaker with a spring constant $k = 341$ N/m. The outer wall is transparent Plexiglas; the bottom plate contains a force sensor flush with its smooth surface. We use 0.430 kg ($\sim 71\,200$) of white spheres of diameter $d = 2.29$ to 2.39 mm, density 0.90 g/cm^3, coefficient of restitution 0.7 ± 0.2, and coefficient of friction 0.3 ± 0.1. The shear plate surface is a densely packed, disordered layer of identical black particles.

The parameter space is large: shearing rate Ω for the upper ring, vibration amplitude A and frequency f, peak acceleration $\Gamma \equiv A(2\pi f)^2/g$, packing fraction ϕ, height H, and compressive load L. We set Ω and Γ at fixed f (60 Hz) and particle number; ϕ varies freely. We use Ωs of 0.010 to 1.6 Hz (nondimensional rate $\tilde{\Omega} \equiv \Omega R/\sqrt{gd} =$ 0.058 to 9.3) and peak accelerations $\Gamma = 0$ to 7. A is small compared to d, e.g., $A = 0.2d$ for $\Gamma = 7$. Relative humidity was 31% to 35% for all runs. We measure forces on a circular plate (diameter 5.4d) centered on the bottom of the cell using a capacitive technique to allow for high speed ($\approx$ kHz) and high sensitivity ($\approx 10^{-5}$ N). As the bottom plate moves in response to both the shaker and the particles, a laser position sensor measures its position and a piezoelectric accelerometer measures Γ. Error in the bottom plate position is ± 0.03 mm, and in H is 0.2 mm.

The system crystallizes when sufficiently vibrated; e.g., sustained vibration at $\Gamma \gtrsim 6$ creates a hexagonally close

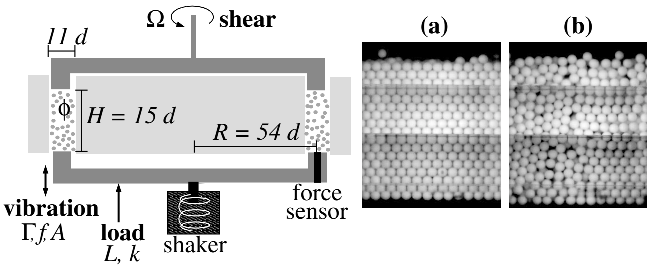

FIG. 1. Schematic cross section of experiment (not to scale). Sample images at outer wall, showing (a) crystallized and (b) disordered phases.

packed (HCP) state with <10 grain boundaries and point defects (holes) in the outer layer. We refer to this state as *crystallized* [9]. Examination of the top surface suggests that order persists throughout the whole cell. Weaker vibration results in other HCP orientations and packings in some regions, as well as disordered regions. To set the cell height, we prepared a fully crystallized state; at $\Gamma = 0$ we then positioned the shearing wheel with only a few particles contacting the particle surface [as in Fig. 1(a)]. Then, the load L has its minimum value, L_{min}. We performed experiments with $L_{\mathrm{min}} \approx 0$; for $L_{\mathrm{min}} > 0$ crystallization by vibration was suppressed.

In contrast, shearing with the upper plate has a disordering effect which is most pronounced near the top of the cell and depends on the parameter regime. At $\Omega = 1.61$ Hz and moderate $\Gamma = 2.0$ the system assumes a *disordered* state. At the beginning of each run, we prepare such a state before setting the desired (Ω, Γ) after which we observe transients of up to five hours. Changes in H are not necessarily monotonic and some states never settle to a characteristic mean H, as discussed below.

Figure 2 shows the hysteretic transition between steady disordered and crystallized states. Downward steps in Ω from a prepared disordered state, cause a decrease in H varying as $\log\Omega$ [10]. A first-order phase transition to the crystallized state occurs at $\tilde{\Omega}_c = 0.2$. Further decreases in Ω result in only slight additional compaction. When we increase Ω from this crystallized state, the transition to disorder is at $\tilde{\Omega}_h = 0.5$, with a return to the dilated state; the total change in H corresponds to 5% of the cell height. While this is a small change, previous 2D studies [11] indicate that a 4% decrease in ϕ is sufficient to cross from a state with a strong force chain network to a fluidlike state without force chains.

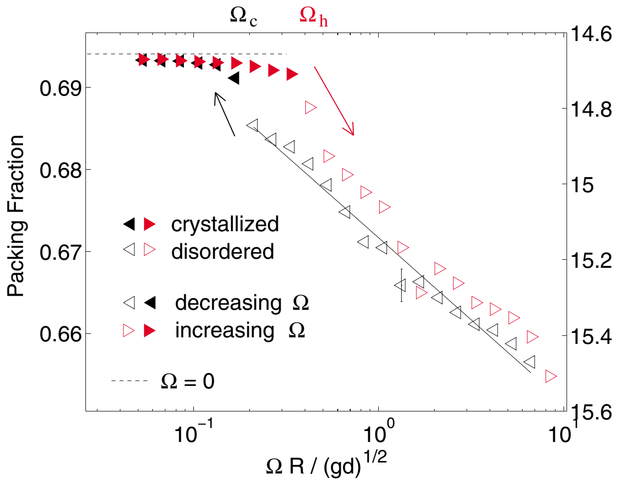

FIG. 2 (color online). Hysteretic transition from disordered (right) to crystallized (left) state for $\Gamma = 2.0$. The dashed line is compaction achieved for $\Omega = 0$ Hz, for comparison. The minimum ϕ does not agree with $\phi_{HCP} = 0.74$ due to the curved geometry and error in the measurement of ball and cell dimensions. The vertical bar represents standard deviation of height measurements.

For $\Gamma = 2.0$, we measured the degree of order vs Ω via 2D spatial autocorrelation functions of images obtained at the outer wall (see Fig. 3). The magnitudes of the off-axis peaks change sharply at Ω_c. There is some order present even for $\Omega > \Omega_c$, due to clusters, particularly in the bottom half of cell. Correlations in the direction parallel to the shear, corresponding to linear "trains" of particles, remain even as the transverse correlation disappears during the shear melting for $\Omega > \Omega_c$. Such trains have been observed as a dominant feature in separate experiments under larger confining pressures [8]. The importance of clusters in the stability of granular systems has been much investigated [12], and further work is necessary in this system.

In addition to the visible changes in ϕ and ordering, there are changes in the forces as well. Probability distribution functions (PDFs) of the forces measured at the bottom of the cell are markedly different for the two states. As shown in Fig. 4, for $\Omega < \Omega_c$ the PDFs are double peaked, corresponding to the two peaks in the PDF of a sinusoid. Thus, we infer that the crystallized particles respond to the vibration of the bottom plate as a solid body. Time traces of the force for $\Omega > \Omega_c$ show amplitude disorder in the force response, but a well-defined frequency. The corresponding PDF is single peaked and the tail of the distribution decays approximately exponentially, a shape characteristic of granular materials [3,13]. The PDFs for increasing and decreasing Ω show the same transition, but at Ω_h and Ω_c, respectively.

As measured from the kurtosis, the PDF is increasingly wide as $\Omega \rightarrow \Omega_c$ (see Fig. 4). This is reminiscent of the large fluctuations near a jamming transition in sheared colloids [14], the broadening of the force PDF for simulations of sheared Lennard-Jones particles [15,16], and the divergence of the viscosity in classical glass transitions. While a crystallized state is fundamentally different from a jammed one due to the presence or absence of order, they share a lack of freedom to rearrange.

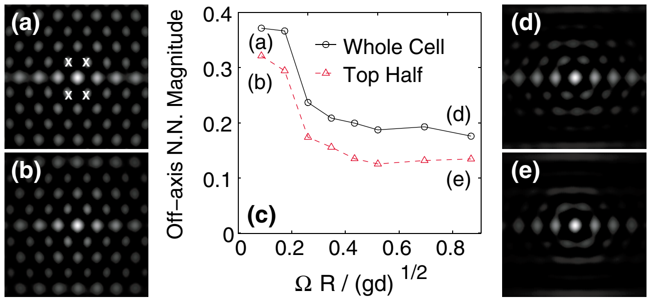

FIG. 3 (color online). Spatial autocorrelation functions of outer wall images for $\Gamma = 2.0$ and values of Ω as indicated in (c), for either the top half of the cell (b),(e) or the whole cell (a),(d). Center (c): The average magnitude of the four off-axis nearest neighbor peaks, marked × in (a), for example. Data collected at decreasing Ω from disordered state.

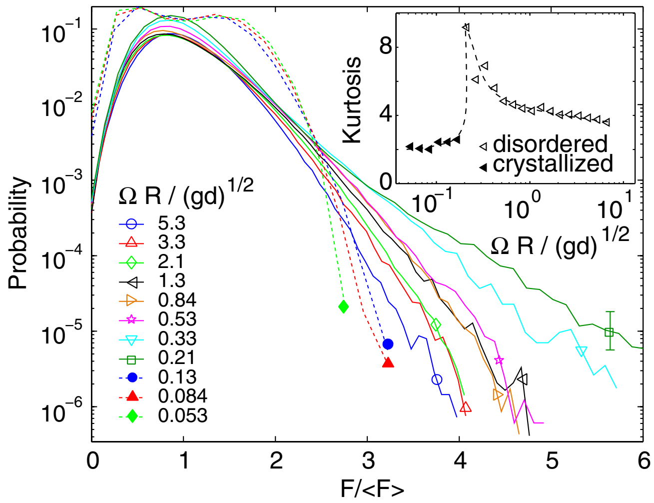

FIG. 4 (color online). Force PDFs (scaled by mean) for representative decreasing-Ω ($\lhd$) points in Fig. 2. Solid lines are disordered and dashed are crystallized. Inset: Kurtosis of the PDFs, showing divergence at Ω_c. The dashed line is a guide to the eye.

As is common with sheared granular materials, the system exhibits an approximately exponential velocity profile localized to the shearing wheel, with a decay length of a few ball diameters. This effect is most extreme in the crystallized state, where the system exhibits stick-slip behavior for the few balls in contact with the shearing wheel, similar to that observed in granular friction experiments [17]. Even in the disordered state, the vibrations do not create a more liquidlike state characterized by an approximately linear velocity profile.

The autocorrelation functions in Fig. 3 show that disorder occurs throughout the cell for $\Omega > \Omega_c$. This full-depth disordering stands in contrast to the disordering localized to the shear surface in molecular dynamics simulations of a 2D shear cell with constant-P boundary conditions and gravity, but without vibration [18]. Since these simulations showed no disordering below the shear band, it appears that vibrations augment the disordering process. The effect we observe could be due to either the fluctuations induced by the vibration itself, or the periodically increased contact with the shearing surface. An initially crystallized state sheared without vibrations fails to disorder the lower portion of the cell, indicating that melting is suppressed. The vibrations therefore act subtly: promoting crystallization at low Ω, but also allowing low-shear areas of the cell to disorder at high Ω.

The crystallization or disordering transition can be understood in terms of the relevant energy scales. Ignoring the effects of a compressive pressure (L is small at Ω_c), shearing and vibration are the only two competing effects. For each, there is an associated kinetic energy input for a particle of mass m: $E_\Omega = \frac{1}{2}m(R\Omega)^2$ and $E_\Gamma = \frac{1}{2}m(2\pi f A)^2$. (These two modes are also anisotropic: the shear acts perpendicular to gravity and the vibration parallel.) For $E_\Gamma > E_\Omega$ the system is crystallized and for $E_\Gamma < E_\Omega$ the system is disordered. The agreement of this line with experimental observations is shown in Fig. 5. There is hysteresis in the state, cell height, and pressure at the line $E_\Gamma = E_\Omega$.

The pressure $P(t)$ on the force sensor shows significant fluctuations, but $\bar{P}(V)$, the average [from $F(t)$] calculated by binning the data by volume V, is a smooth function, although not necessarily linear. Changes in P are linked, through the effective spring constant of the shaker, to changes in V. The slope dP/dV corresponds to a value less than that associated with the spring constant, presumably because the vertical pressure is redirected to the sidewalls (Janssen effect).

For runs in the disordered state ($\tilde{\Omega} = 0.27$), we observe a linear $P(V)$ when varying $\Gamma < \Gamma_c$. In these experimental runs, the system consists of coexisting, fluctuating regions of partial disorder and crystallization while still remaining along this characteristic line. (See open triangles and squares in Fig. 6(a) and inset.) The system can become almost entirely crystallized before disordering (dilating), but can also permanently end up in the fully crystallized state. The system displays similar intermittency when started from an over-dilated state and allowed to evolve to a steady state: during net compaction, the system routinely redilates slightly before compacting further. Furthermore, we observed the same slope, but offset in P, during runs with higher L_{min} or humidity. As shown in Fig. 6(b), for $\Gamma = 2$ we observe the same $P(V)$ for low $\tilde{\Omega}$, and the remaining runs map out a continuous function $P(V)$ with smaller dP/dV at higher Ω.

Significantly, the system's pressure and height change dramatically in exploring $P(V)$, a phenomenon tied to large-scale fluctuations in the relative amounts of disor-

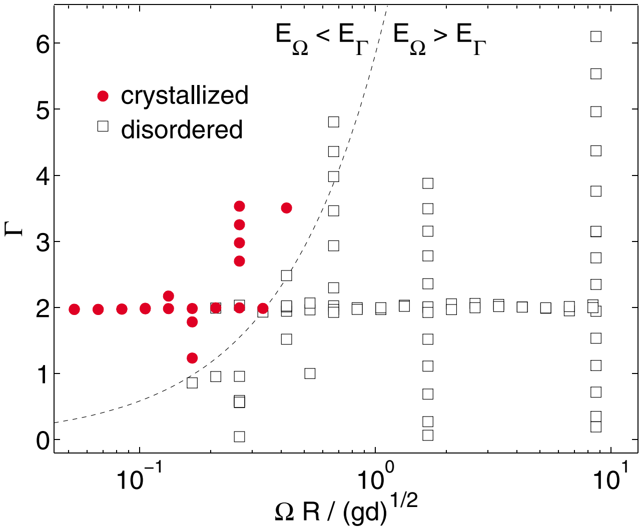

FIG. 5 (color online). Phase diagram in Ω and Γ. Observations of crystallized (●) and disordered state (□). Overlapping circles and squares are hysteresis from Fig. 2. The dotted line is $E_\Omega = E_\Gamma$.

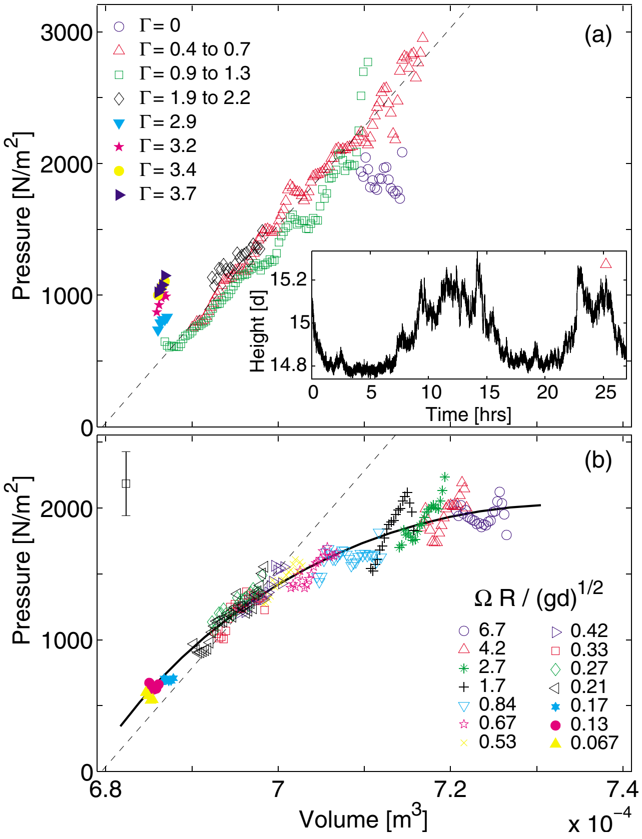

FIG. 6 (color online). Dependence of $\bar{P}$ on V; solid symbols are crystallized states. (a) $\Omega = 0.051$ Hz ($\tilde{\Omega} = 0.27$) and varying Γ; the inset shows intermittency in a run marked △. (b) $\Gamma = 2.0$ and varying Ω. The solid line is a guide to the eye; the dashed line is fit to (a), shown also on (b) for comparison.

dered vs ordered regions. It is remarkable that at lower ϕ the system sustains higher forces. While this is a natural consequence of Reynolds dilatancy, whereby shearing causes an increase in V, the underlying process remains to be understood. One interpretation is that points along the curve $P(V)$ vary according to an order parameter, X, describing the state of the system. Such a parameter may represent the degree of partial disorder or the strength of the force chains. Thus, we write $(dP/dV) = (\partial P/\partial V)_X + (\partial P/\partial X)_V(dX/dV)$, and note that $\partial P/\partial V$ is negative as for ordinary materials. Therefore, the second term must be positive and greater than $\partial P/\partial V$ to account for $dP/dV > 0$, as required by the spring-loaded boundary condition.

In this Letter, we have described competing disordering and crystallizing effects for sheared/vibrated granular materials. While the concept of a granular temperature has provided a thermodynamic analog for granular gases, the findings here point to several caveats regarding that association. External vibration fails to provide "heating" in a thermodynamic sense: the resulting fluctuations do not allow the shear profile to be liquidlike, and they produce crystallization rather than melted disorder. However, fluctuations introduced by the shearing of the material have the opposite effect. Future work should be undertaken to examine temperaturelike (or other) variables which might control this phase transition.

Granular materials are often studied with constant-volume constraints, where they exhibit higher forces for higher packing fractions. In contrast, by allowing dilation, these experiments allow the system to explore a range of pressure and volume states. It is interesting that $\bar{P}$ takes its unique value from both V and Ω (and, to a lesser extent, Γ) for the given spring constant. While Γ is crucial to determining the ordered or disordered state, within the disordered state $\bar{P}$ is essentially independent of this parameter. Since changes in Ω or Γ induce changes in the order or disorder as well as controlling $\bar{P}$ (see Figs. 3 and 6), one possible interpretation of the data is the existence of an order parameter which controls the transition. Also of interest is the novel logarithmic dependence of the packing fraction on Ω.

The authors would like to thank Lou Kondic, Jerry Gollub, and Matthias Sperl for fruitful conversations. This work has been supported by the NASA microgravity program, Grant No. NNC04GB08G.

[1] B. J. Ackerson and N. A. Clark, Phys. Rev. Lett. **46**, 123 (1981).
[2] B. J. Ackerson and P. N. Pusey, Phys. Rev. Lett. **61**, 1033 (1988).
[3] B. Miller, C. O'Hern, and R. P. Behringer, Phys. Rev. Lett. **77**, 3110 (1996).
[4] W. Losert *et al.*, Phys. Rev. Lett. **85**, 1428 (2000).
[5] D. M. Mueth *et al.*, Nature (London) **406**, 385 (2000).
[6] D. Helbing, I. J. Farkas, and T. Vicsek, Phys. Rev. Lett. **84**, 1240 (2000).
[7] E. R. Nowak *et al.*, Powder Technol. **94**, 79 (1997).
[8] J. C. Tsai, G. A. Voth, and J. P. Gollub, Phys. Rev. Lett. **91**, 064301 (2003).
[9] Interestingly, the crystallization improves when the vibration is supplemented by weak shear ($\Omega \approx 10^{-2}$ Hz).
[10] R. R. Hartley and R. P. Behringer, Nature (London) **421**, 928 (2003).
[11] D. Howell, R. P. Behringer, and C. Veje, Phys. Rev. Lett. **82**, 5241 (1999).
[12] G. D. R. Midi, Eur. Phys. J. E **14**, 341 (2004); T. G. Drake, J. Geophys. Res. **95**, 8681 (1990); W. Polashenski *et al.*, Phys. Rev. E **66**, 021303 (2002); M. L. Tan and I. Goldhirsch, Phys. Fluids **9**, 856 (1997); G. Chambon *et al.*, Phys. Rev. E **68**, 011304 (2003).
[13] D. M. Mueth *et al.*, Phys. Rev. E **57**, 3164 (1998).
[14] D. Lootens, H. Van Damme, and P. Hebraud, Phys. Rev. Lett. **90**, 178301 (2003).
[15] C. S. O'Hern *et al.*, Phys. Rev. Lett. **86**, 111 (2001).
[16] J. H. Snoeijer *et al.*, Phys. Rev. Lett. **92**, 054302 (2004).
[17] S. Nasuno *et al.*, Phys. Rev. E **58**, 2161 (1998).
[18] P. A. Thompson and G. S. Grest, Phys. Rev. Lett. **67**, 1751 (1991).