SPECIAL TOPICS

Physics of Soft Condensed Matter

Granular Gases in Compartmentalized Systems

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Although both granular gases (GG) and molecular gases (MG) are characterized by random motions of their constituents, phenomena not possible for MG, such as clustering and Maxwell's demon are reported in GG. The origin of these intriguing phenomena is the dissipative collisions in GG which are coupled to the local density of the GG in a spatially extended or compartmentalized system. Systems with two or more types of grains are especially interesting because the asymmetry in the dissipative collisions between different types of grains can lead to oscillations and even more interesting dynamics. In this article, flux models with different granular temperatures for different types of grains are studied to understand and explore these phenomena.

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1. Introduction

Traditionally, properties of granular materials would only be interested to engineers who mainly concern their static mechanical properties for practical reasons.¹⁾ However, the dynamics of granular materials turn out to be extremely rich and have attracted attentions from physicists of different fields.²⁻⁴⁾ Some of the famous examples are the heap formation of a granular bed^{5-7} and the size segregation of a granular system with grains of various sizes under vertical vibration known as the Brazil Nut effects.^{8,9)} In these phenomena, energy is being injected continuously into the system by the oscillating boundaries and propagated into the bulk by the inelastic collisions of the grains. A steady state of the whole system is reached when the dissipation of the system is balanced by the input of the energy. Since both the energy input and dissipation depends crucially on the configurations of the system, many intriguing steady states and even oscillatory states can be created.

In both the heap formation and the Brazil nut effects, the density of the system is high and the agitated granular system looks like a liquid. Therefore, concepts such as convection and buoyancy sometimes are used to explain the observed phenomena. In the case of low density such as a granular gas, one might naturally borrow concepts from their molecular counter parts. At first sight, granular gases (GGs) and molecular gases look similar, both are characterized by random motions and collisions. Similar to the temperature of a molecular gas, the concept of granular temperature¹⁰ is sometimes found to be useful. However, the GGs are only in a steady state in which input and loss of energy are being balanced. They are not in thermal equilibrium as their molecular counter parts and the laws of thermodynamics for molecular gases do not apply for the GGs. For example, the

thermodynamically impossible phenomenon such as the Maxwell's demon^{11,12}) has been produced and successfully explained. In such an experiment, a GG confined in a compartmentalized system (see Fig. 1) can be induced to segregate into only one of the compartments by lowering the vibration amplitude of the system. In this latter case, a decrease of the configurational entropy of the system takes place spontaneously; as if the second law of thermodynamics is violated. In fact, other similar intriguing segregation¹³ and ratchet effects¹⁴ have also been reported in compartmentalized granular gases.¹⁵

Since GGs are only in a quasi-steady state of energy balance, many different configurations of the system are possible, the transitions between these possible states give rise to the intriguing pattern formation and dynamics in granular systems under vibration; especially for a spatially extended system. In a spatially extended system, it is possible that different parts of the system are in different configurations in which local energy balance is still achieved. For example, for the heap formation, most of the grains in a uniformly distributed layer of granular materials will be concentrated in one corner of the container to form a heap under sufficiently strong vibration. One can identify the grains in the heap as in a condensed phase of the granular gas while the very few number of grains outside the heap as in the gas phase. The grains in the gas phase will have higher velocities than those in the condensed phase because they have direct contact with the vibrating boundary and little inter-particle collisions. However, for the grains in the condensed phase, because of the high density, energy gained from the collisions with the boundaries is rapidly dissipated by the inter-grain inelastic collisions in the bulk. If one uses the concept of granular temperature which is related to the kinetic energy of the grains, one will find that the grains in the heap are at a lower temperature than those outside the heap. Clearly, one can see that a "temperature" gradient is thus created by the external vibrations in a spatially extended system because

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of the self-organization of the spatial distribution of the grains.

Obviously, the above situation becomes more interesting when there are more than one type of grains in a spatially extended system. For example, because of the asymmetry in the dissipative collisions between different types of grains, the energy injected from the vibrating boundary will be distributed differently to different types of grains. Therefore, different types of the grains can have different granular temperatures at the same time; even at the same location in the system. Presumably, the self-organization of these grains in a spatially extended system leads to the many intriguing dynamics reported recently. In this article, we will briefly review some of these phenomena and discuss the case of granular oscillation in details in terms of a flux model with different granular temperatures for different types of grains.

2. Maxwell Demon: Clustering of Grains in Compartmentalized Chamber

We will start with the case of the Maxwell's demon. Filling N plastic particles in a box separated into two identical compartments, connected by a narrow horizontal slit at a finite height h, Schlichting and Nordmeier¹¹⁾ in 1996 performed an experiment on dilute gas of granular materials by vertical vibration. When the shaking strength is high enough, particles will distribute equally to both sides of the box. As the vibration strength is lowered than a certain critical value, this symmetry in population will be broken, and particles will settle preferentially on one side of the box. The clustering of grains into one side of the system is referred as the "Maxwell's demon" phenomenon. The particles in the connected two compartments are at different temperatures: one at dilute phase of high temperature and the other at dense phase of low temperature. Eggers in 1999 developed a flux model¹² and obtained a quantitative theory for this clustering phenomenon. It was found in good agreement with his numerical simulation results. In 2001 Lohse's group studied the vibro-fluidized granular gas in 2 and 3 compartmentalized containers.¹³⁾ They found that for sufficiently strong shaking the population is equi-partitioned, but when the shaking intensity is lowered, the gas clustered in one compartment. The phase transition towards the cluster state is of 2nd order for 2-compartments and 1st order for 3-compartments. This clustering effect with 3 or more compartments is found to be hysteretic.^{13,16} Later in 2001 Brey et al.¹⁷) reported observations of spontaneous symmetry breaking in simulations of a vibrated system confined in two connected compartments under no gravity, and the phase transition was satisfactorily described by hydrodynamic equations.

As discussed in the previous section, granular systems, in which ordinary thermal fluctuations do not play a role, often exhibit various ordered patterns. Patterns form when the systems turn out to be in multiple meta-stable steady states, which are far from equilibrium. Spatial ordered structures, such as regular surface patterns, oscillons, segregation etc., have been observed in different granular systems.¹⁸ GG in compartmentalized system showed a variety of patterns far from equilibrium, with the Maxwell demon clustering being a classic example. A typical setup for such a system is shown in Fig. 1, which can manifest the Maxwell demon phenom-



Fig. 1. (Color online) A schematic of a typical setup for a granular gas in a compartmentalized chamber under vertical vibrations. The temperature and number of grains in the left (right) chamber are denoted by T_L (T_R) and N_L (N_R) respectively.

enon. Figure 1 is a 2-compartment system which is made up of a right compartment (RC) and a left compartment (LC). Hereafter, quantities in the LC and RC will be denoted by the L and R subscripts respectively.

To realize such a demon state by dint of dissipation, most attention had been given to the clustering phenomenon of mono-disperse granular gases in this system.^{12,17,19,20} And this granular demon state was easily observed, while the clustering dynamics and the bifurcation instabilities were well explained by several flux models,²¹⁾ which described the flows of particles between the compartments. Here we illustrate that this clustering or "Maxwell demon" phenomenon can be understood as an unstable evaporation and condensation unique to granular systems. For a single type of grains (number of grains, N, mass, m, radius, r, coefficient of resititution, e), under vertical vibration of a bottom plate of area Ω and speed v, by balancing the energy input rate due to the vibrating bottom plate ($\simeq mgNv$ for $v \ll \sqrt{k_{\rm B}T/m}$ to the dissipation due to inelastic collisions among the grains $[=4gN^2r^2(1-e^2)\sqrt{\pi mk_{\rm B}T}/\Omega]$, one can easily derive the steady state granular (kinetic) temperature, as

$$k_{\rm B}T = \frac{m}{\pi} \left[\frac{v\Omega}{4(1-e^2)r^2N} \right]^2 \tag{1}$$

where $k_{\rm B}$ is the Boltzmann constant. Such a relation can be intuitively understood from the picture that the kinetic temperature of a grain comes from the energy of vibrating plate ($\propto v^2$) and shared by the binary inelastic collisions ($\propto 1/N^2$). The key idea in explaining Maxwell demon is that the granular temperature of the grains is given by $T \propto (v/N)^2$ which constitute the mechanism for the instability that gives rise to clustering of monodispersed grains into one side of a compartmentalized chamber.^{12,22)}

For a mono-disperse granular gas in a single compartment, there is a single granular temperature for the system, T, which is inversely proportional to N^2 because most of the dissipation of the system comes from the binary collisions of the grains among themselves and higher density means more collisions and therefore a lower T. However, for the case as shown in Fig. 1 with two chambers, when v is large, one would expect that the grains are free to move back and forth between LC and RC. Therefore, the grains are distributed equally ($N_R = N_L = N/2$) in LC and RC with $T_R = T_L$. When v is lowered to a point that most of the grains in RC and LC cannot be exchanged, the system can be unstable. Consider a fluctuation of ΔN in the number of grains in the RC, i.e., $N_{\rm R} = N/2 + \Delta N$, $N_{\rm L} = N/2 - \Delta N$. In this case, $T_{\rm L}$ will be higher than $T_{\rm R}$ and more grains will jump from LC to RC than those from RC to LC. During this process, $T_{\rm L}$ is raised further by the evaporation of grains and *vice versa* for the condensation in RC. Therefore this process is unstable and it will not stop until the difference in grain number between the two compartments is so large that the smaller probability for grains to jump from RC to LC is compensated by a large number of $N_{\rm R}$.

3. Binary Mixture of Grains in a Two-Compartment Chamber

Bidisperse GG in compartmentalized system was first studied experimentally in 2002 by Lohse's group²⁰⁾ using steel balls which experimentally showed the competitive clustering phenomenon between the two compartments. The radius ratio of the large and small beads is fixed as 2:1 and the number ratio is 1:2. The chamber is mounted on a shaker with adjustable amplitude A and frequency f. The inverse shaking strength $1/(Af)^2$ is found to be one of the crucial parameters for the clustering behavior. They found that depending on the shaking strength, the clustering can be directed either towards the compartment initially containing mainly small particles or to the one containing mainly large particles. And they generalized Eggers' flux model¹²⁾ to bidisperse GGs to explain what they observed. This experimental finding was carefully studied and reported in 2004 by Lohse's group with MD simulation and theoretical modeling.²³⁾ Though no oscillatory state was found either in their experiments nor numerical simulation results, two later papers predicted this so-called granular clock phenomenon in two-dimensional (2D) systems, through molecular dynamics simulations and theoretical models: Costantini et al.²⁴⁾ simulated smooth hard disks with equal sizes but different masses, and developed a mean field theory; while Lambiotte et al.25) simulated those with different sizes but equal masses, and suggested a four-dimensional dynamical system description. Experiments in ref. 26 showed the existence of such an oscillation with large and small glass beads in a quasi-2D system, and Lambiotte et al.'s model was adopted to explain their results in terms of reverse Brazil nut effects. However, the above models were not in good quantitative agreements with experiments nor theoretically satisfactory. Our latest results²²⁾ showed the oscillation of glass and steel balls in a three-dimensional system, and a new phenomenological model was proposed to show the instability of the equi-partition state is via a supercritical Hopf bifurcation giving rise to oscillation.

3.1 Granular clock: Brief review of experimental results

Here we briefly review some experimental results for binary mixtures of grain in a two-compartment system. Miao *et al.*²⁷⁾ first observed oscillatory phenomena in some preliminary experiments using millet and mung beans, which were different in both sizes and masses. However, they claimed no oscillation for large and small beads made from the same material. Markus' group in 2006 reported²⁶⁾ their observations of oscillations and full segregation in a quasi 2-D system containing two compartments of bidisperse granular gas consisting of particles with equal densities but different diameters or grains of equal diameters but different densities. Soda lime glass spheres with 138 smaller spheres (diameter 2 mm) and 27 larger spheres (diameter 4 mm) were used; and for experiments with binary mixtures of particles having equal sizes (diameters: 2 mm) and different densities, 92 spheres of pairs of spheres of polyacetal, glass, steel, and bronze were used. Oscillations between the left and right compartments were observed and were interpreted using mechanism involving reverse Brazil nuts effects which lead to *ad hoc* terms in the dynamical model equations.

The recent experiments performed by our group are carried out in three-dimensional (3D) rectangular cells and with grains of equal sizes.²²⁾ The detailed dynamics and phase diagram was obtained. The experimental setup is consisted of a rectangular glass container of 2.6 cm wide and 5.4 cm long with height of 13.3 cm, divided into two equal compartments by an aluminum wall of 0.2 cm thick. Particles in the two compartments are connected by a window of $28 \times 18 \text{ mm}^2$ on the wall. The bottom edge of the window can be at a height (h) of 0.9, 1.5, and 2.0 cm from the bottom plate. This glass container is mounted on a shaker to bring the system into a gaseous state through vertical, sinusoidal vibrations with adjustable frequency fand amplitude A. Steel balls and glass beads of the same size, 0.5 or 1 mm in radius, are used. Observation of the dynamics of the systems is carried out as a function of the shaking velocity v with $v = 2\pi A f$. In this experiment three regimes, namely, segregation, oscillation and homogeneous distribution, of the two types of grains in the two compartments are observed at different number ratios and shaking velocities v. In the experiments to determine the phase diagram (Fig. 2, the total number of grains is fixed at 960 while the ratio of number of glass beads to that of the steel beads (χ_0) varies from 1 : 1 to 7 : 1. Video and pictures are taken to record the observed particle motions. The boundaries of these regimes are determined by measuring either the oscillation amplitude or its time period. The oscillation amplitude is measured by counting the number of remaining steel particles when maximum particles are in the other compartment. The time period of the oscillation is determined using a timer at the lower boundary as the oscillation is longer than a few seconds to minutes.

Results of our experiments are summarized as a phase diagram in terms of v and χ_0 as shown in Fig. 2 obtained with f = 60 Hz. Three states [homogenous (HS), oscillatory (OS), and segregated (SS)] in the distribution of grains can be observed as v increases. These phases are separated by two transition velocities $v_{\rm f}$ and $v_{\rm c}$ which are functions of χ_0 . It is shown that the boundaries of these three regimes are sensitive only to the combination of $Af \propto v$. For a fixed value of χ_0 , as v is being lowered, the system is first in HS when $v > v_c$ and then turns into OS when $v_c > v > v_f$. The system becomes segregated (SS) when v is lowered below $v_{\rm f}$. The features of the transition points $v_{\rm c}$ and $v_{\rm f}$ are that the amplitude of the oscillations (Δ) increases with ($v_c - v$) while the period of the oscillation (τ) decreases with $(v - v_f)$. More details of the experimental results can be found in ref. 22. The dynamics of the clock is explained in terms of an unstable evaporation/condensation model for the GG in which the temperatures of the two types of grains are



Fig. 2. (Color online) Phase diagram of binary mixture of steel and glass beads under vibration.

considered to be different and they are functions of the composition of the GG. In our model, oscillations in the system are driven by the asymmetric collisions properties between the two types of grains. Some details of the model will be presented in the following section.

3.2 Dual temperatures in binary mixture of grains

To understand the observed oscillations in experiments, it is essential to realize that two different temperatures exist for the two types of grains in the binary mixture. Consider now the simpler case of a chamber (not compartmentalized) consists of two type of grains A and B, each with masses and radii m_{α} and r_{α} ($\alpha = A$ or B), and the total number of grains are N_A and N_B respectively. The upward vertical direction is the z-axis and the number density of grain is denoted by $n_{\alpha}(z)$. Due to gravity (denoted by g), the grain density follows the barometric distribution given by

$$n_{\alpha}(z) = n_{\alpha}(0) \exp\left(-\frac{m_{\alpha}gz}{k_{\rm B}T_{\alpha}}\right) \quad \alpha = A \text{ or } B.$$
 (2)

 $n_{\alpha}(0) = (m_{\alpha}gN_{\alpha})/(\Omega kT_{\alpha})$ which follows from the normalization condition $\int_{0}^{\infty} n_{\alpha}(z) dz = N_{\alpha}/\Omega$. If the initial speeds of A and B grains are $u_{\rm A}$ and $u_{\rm B}$ respectively, one can get the kinetic energy change of A-grain in an AB collision, $\Delta \mathcal{K}_{\rm A}$, by direct calculation. The energy change rate/volume of A-grains due to AB collision is

$$q_{\rm A}^{\rm (AB)} = n_{\rm A}(z)n_{\rm B}(z)\pi(r_{\rm A}+r_{\rm B})^2 \langle |\boldsymbol{u}_{\rm A}-\boldsymbol{u}_{\rm B}|\Delta\mathcal{K}_{\rm A}\rangle.$$
 (3)

The mean dissipation rate of A-grain due to AB collision is obtained by $Q_A^{(AB)} = -\Omega \int_0^\infty dz \, q_A^{(AB)}$. Then after some algebra, one finally obtains²⁸

$$Q_{\rm A}^{\rm (AB)} = -\frac{2\sqrt{2\pi m_{\rm A}m_{\rm B}g}}{\Omega(m_{\rm A} + m_{\rm B})^2} (r_{\rm A} + r_{\rm B})^2 \times N_{\rm A}N_{\rm B}(1 + e_{\rm (AB)})(e_{\rm (AB)}m_{\rm B} - m_{\rm A})\sqrt{\sigma_{\rm A}^2 + \sigma_{\rm B}^2},$$
(4)

where $e_{\alpha\beta}$ denotes the restitution coefficient between the α type and β -type grains ($\alpha = A$ or B) and $\sigma_{\alpha} \equiv \sqrt{k_{\rm B}T_{\alpha}/m_{\alpha}}$. Thus A-grains can gain energy from AB collisions (i.e., Bgrains heat up A-grains) if $e_{(AB)}m_{\rm B} > m_{\rm A}$. The dissipation rate of A-grains due to A–A collisions can also be computed in a similar way to give

$$Q_{\rm A}^{({\rm AA})} = rac{4\sqrt{\pi}m_{\rm A}g}{\Omega}r_{\rm A}^2N_{\rm A}^2(1-e_{({\rm AA})}^2)\sigma_{\rm A}.$$

Thus the total dissipation rate of A-grains is given by $Q_A^{(AB)} + Q_A^{(AA)}$ which would be equal to the mean energy input rate (denoted by J_A) from the bottom vibrating plate colliding with the A-grains. Similar expressions for the dissipation rates of B-grains due to AB collisions ($Q_B^{(AB)}$) and BB collisions ($Q_B^{(BB)}$) can be similarly obtained simply by interchanging A \leftrightarrow B in the expressions for $Q_A^{(AB)}$ and $Q_A^{(AA)}$ respectively.

On the other hand, the rate of energy input due to collisions between the vibrating bottom plate and the grains is $J_{\alpha} \simeq m_{\alpha}gN_{\alpha}v$ ($\alpha = A$ or B). For a binary mixture of A and B grains under steady condition, the energy input rate of each type of grain is balanced by its total dissipation rate, namely $J_{\rm A} = Q_{\rm A}^{({\rm AB})} + Q_{\rm A}^{({\rm AA})}$ and $J_{\rm B} = Q_{\rm B}^{({\rm AB})} + Q_{\rm B}^{({\rm BB})}$. One can obtain,²⁸⁾ after some algebra,

$$\sigma_{\rm A} = \frac{\upsilon}{D_{\rm A}N_{\rm A}} + \frac{\eta}{D_{\rm A}\chi} \left(\frac{e_{\rm (AB)}m_{\rm B}}{m_{\rm A}} - 1\right) \sqrt{\sigma_{\rm A}^2 + \sigma_{\rm B}^2}$$
$$\sigma_{\rm B} = \frac{\upsilon}{D_{\rm B}N_{\rm B}} - \frac{\eta\chi}{D_{\rm B}} \left(1 - \frac{e_{\rm (AB)}m_{\rm A}}{m_{\rm B}}\right) \sqrt{\sigma_{\rm A}^2 + \sigma_{\rm B}^2} \tag{5}$$

where

$$\chi \equiv N_{\rm A}/N_{\rm B},$$

 $D_{\rm A} \equiv \frac{4\sqrt{\pi}r_{\rm A}^2(1-e_{\rm (AA)}^2)}{\Omega}$ (*D*_B is similarly defined)

and

$$\eta \equiv 2\sqrt{2\pi} (1 + e_{(AB)}) \frac{(r_{A} + r_{B})^{2}}{\Omega} \frac{m_{A}m_{B}}{(m_{A} + m_{B})^{2}}$$

 D_A and D_B can be interpreted as dissipation factors because they vanish in the limit of elastic collisions. However, such a limit is not relevant in our model in which dissipations are always important. It is easy to see that the case of a single temperature, i.e., $T_A = T_B$ or $\sigma_A = \sigma_B \sqrt{m_A/m_B}$, is in general not a possible solution to eq. (5). Hence the coexistence of two temperatures for two different grain types is a generic phenomenon due to the asymmetric properties of the grain types. Furthermore, by directly iterating eq. (5), one can readily see that the solutions for σ_A and σ_B can be put into the forms

and

$$\sigma_{\rm A} = \frac{v}{D_{\rm A} N_{\rm A} p(\chi)}$$

$$\sigma_{\rm B} = \frac{v}{D_{\rm B} N_{\rm B} q(\chi)},$$

for some function p and q depending on the parameters. Only the dependence on χ is explicitly written to emphasis the most crucial parameter in this problem. Figure 3(a) shows typical behavior of $p(\chi)$ and $q(\chi)$ as solved from eq. (5). $p(\chi)$ approaches to 1 for large χ , whereas $q(\chi)$ increases with χ . In ref. 22, it was assumed the functional forms of $p(\chi) = \{2 - [1/(1 + \chi)]\}/2$ and q = 2p for numerical solutions of the flux model. Here we can examine how good is such an assumption. From our theoretical result, pand q are not trivially related by a constant factor and thus the assumption of q = 2p is not good quantitatively, but it



Fig. 3. (Color online) (a) Theoretical calculations of the functions $p(\chi)$ and $q(\chi)$. $e_{(AA)} = 0.35$, $e_{(AB)} = 0.30$, $e_{(BB)} = 0.25$, and $m_A/m_B = 0.25$. (b) Theoretical results of the functions $p(\chi)$ for different mass ratios of the binary GGs. Value of restitution coefficients are the same as in (a). The approximated functional form of $p(\chi) = \{2 - [1/(1 + \chi)]\}/2$ (solid curve) is plotted showing that it gives good qualitative description.

does give qualitative correct features of the system. On the other hand, the assumed form of $p(\chi) = \{2 - [1/(1 + \chi)]\}/2$ does provide a reasonable approximation to the actual $p(\chi)$ from our calculation. As shown in Fig. 3(b) for $p(\chi)$ for different mass ratios, as least for some range of parameters, the assumed form of $p(\chi)$ is rather good. The functions $p(\chi)$ and $q(\chi)$ can also be measured experimentally and these results will be reported elsewhere.²⁸⁾

3.3 Temperature oscillations and flux model

Let us now consider the case as shown in Fig. 4 with two different types of grains: A and B. For such a bi-disperse gas in a two compartment system, relevant parameters are: $N_A = N_{AL} + N_{AR}$, $N_B = N_{BL} + N_{BR}$, $\chi_L = N_{AL}/N_{BL}$, and $\chi_R = N_{AR}/N_{BR}$. For high v, we will have: $N_{AL}/N_A =$ $N_{BL}/N_B = 1/2$ (i.e., $\chi_L = \chi_R = \chi_0$), $T_{AL} = T_{AR}$, and $T_{BL} =$ T_{BR} with $T_B < T_A$. When the strength of shaking is lowered, one will come to a point at which only A-grains are free to exchange between LC and RC. When this happens, the system is similar to that of a single type of A, and our arguments about clustering will be applicable. That is: there will be an unstable evaporation and condensation of A (upper left in Fig. 4).

However, the situation here is more complicated because there will be changes in χ_L and χ_R during this process. Figure 4 illustrates schematically the situation. When grains A evaporate from RC and condense into LC, both $T_{\rm AL}$ and $T_{\rm BL}$ are lowered while those in RC will be raised. When enough of grains A are evaporated from RC, the T_{BR} will be raised and it can be so high that the grains B start to evaporate from RC and condense in LC too (upper right in Fig. 4). Since this is an unstable situation, once it happens, more of grains B will jump from RC to LC. However, when grains B start to jump from RC to LC, T_{AL} starts to rise too. When enough B has jumped from RC to LC, T_{AL} will be so high that the grains A start to jump from LC back to RC (lower left in Fig. 4). Again, this is an unstable situation which will lead to most of the grain A to jump from LC to RC (lower right in Fig. 4). That is: there is an oscillation in



Fig. 4. (Color online) Schematic diagrams showing the phenomenon of temperature oscillation in a two-compartment system with a bi-disperse granular system. Temperature or grain oscillation of A-grains is illustrated in the following time sequence: Upper left: fluctuations cause the evaporation of A-grains from RC to LC. Upper right: T_{BR} becomes higher and unstable evaporations of B in RC begins. Lower left: T_{AL} becomes higher and unstable evaporations of A in LC begins. Lower right: most A-grains are now in the RC, T_{BL} becomes higher and B grains can evaporate and jump from LC to RC. See text for details.

grains A. Similar arguments will show that there will also be oscillations in grains B too.

Quantitative flux model can be constructed with the above physical picture. Suppose a slit of area *S* is open at a height *h* on one side of the vertical wall separating the two compartments, the flux of α -type grains ($\alpha = A$ or B) through the slit dN_{α}/dt can be calculated as follows. The horizontal velocity distribution follows a zero-mean Gaussian distribution with variance $\sigma_{\alpha} \equiv \sqrt{k_{\rm B}T_{\alpha}/m_{\alpha}}$, which is also the mean horizontal speed. Thus the flux escaping from the slit is given by $-dN_{\alpha}/dt = S\sigma_{\alpha}n_{\alpha}(h)$, using the barometric distribution in eq. (2), one has

$$-\frac{\mathrm{d}N_{\alpha}}{\mathrm{d}t} = \frac{SgN_{\alpha}}{\sigma_{\alpha}\Omega} \exp\left(-\frac{gh}{\sigma_{\alpha}^2}\right). \tag{6}$$

Since $\sigma_A = v/[p(\chi)N_AD_A]$, hence the flux of A grains leaving the compartment is given by

$$-\frac{\mathrm{d}N_{\mathrm{A}}}{\mathrm{d}t} = K_{\mathrm{A}} \frac{N_{\mathrm{A}}^{2}}{\nu/p(\chi)} \exp\left\{-a_{\mathrm{A}} \left[\frac{N_{\mathrm{A}}}{\nu/p(\chi)}\right]^{2}\right\}$$
(7)

where

$$K_{\rm A} \equiv \frac{SgD_{\rm A}}{\Omega}$$

and

$$a_{\rm A} \equiv ghD_{\rm A}^2 = \frac{h}{g} \left(\frac{K_{\rm A}\Omega}{S}\right)^2.$$

The balance of the fluxes of A and B between RC and LC gives:

$$\frac{1}{K_{\rm A}}\dot{N}_{\rm AL} = -\frac{N_{\rm AL}^2}{v_{\rm A}(\chi_{\rm L})}\exp\left[-a_{\rm A}\frac{N_{\rm AL}^2}{v_{\rm A}^2(\chi_{\rm L})}\right] \\ +\frac{(N_{\rm A}-N_{\rm AL})^2}{v_{\rm A}(\chi_{\rm R})^2}\exp\left[-a_{\rm A}\frac{(N_{\rm A}-N_{\rm AL})^2}{v_{\rm A}^2(\chi_{\rm R})}\right] \\ \frac{1}{K_{\rm B}}\dot{N}_{\rm BL} = -\frac{N_{\rm BL}^2}{v_{\rm B}(\chi_{\rm L})}\exp\left[-a_{\rm B}\frac{N_{\rm BL}^2}{v_{\rm B}^2(\chi_{\rm L})}\right] \\ +\frac{(N_{\rm B}-N_{\rm BL})^2}{v_{\rm B}(\chi_{\rm R})}\exp\left[-a_{\rm B}\frac{(N_{\rm B}-N_{\rm BL})^2}{v_{\rm B}^2(\chi_{\rm R})}\right]$$
(8)

where $a_{\rm B} \equiv ghD_{\rm B}^2$.

With these notations, we have:

and

$$k_{\rm B}T_{\rm B} = m_{\rm B} \left(\frac{v_{\rm B}}{D_{\rm B}N_{\rm B}}\right)^2.$$

 $k_{\rm B}T_{\rm A} = m_{\rm A} \left(\frac{v_{\rm A}}{D_{\rm A}N_{\rm A}}\right)^2$

The two velocities v_A and v_B are related to the shaking velocity as: $v_A = v/p(\chi)$ and $v_B = v/q(\chi)$ for some general functions p and q where $\partial p/\partial N_B < 0$ and $\partial q/\partial N_A > 0$. The requirements on $\partial p/\partial N_B$ and $\partial q/\partial N_A$ are used to implement our assumption that the temperature of grains A is raised by the presence of B while the presence of grains A lowers the temperature of grains B. Obviously, $N_{AL} = N_A/2$ and $N_{BL} =$ $N_B/2$ is a fixed point but it will be stable only when v is large. When v is smaller than a threshold v_c , it will become unstable and turn into a limit cycle. This has been demonstrated in²²⁾ for the assumed functional forms: $p(\chi) =$ $\{2 - [1/(1 + \chi)]\}/2$ and q = 2p by numerically solving eq. (8).

3.4 Hopf bifurcation at v_c

To investigate in more detail analytically the transition from the homogeneous uniform state to the oscillating state, we define the deviation from the fixed point $N_{\rm AL} = N_{\rm A}/2$ and $N_{\rm BL} = N_{\rm B}/2$ by $\epsilon_{\rm A} = N_{\rm AL}/N_{\rm A} - (1/2)$ and $\epsilon_{\rm B} = N_{\rm BL}/N_{\rm B} - (1/2)$, the governing equations can be put into the form

$$\epsilon_{\rm A} = -F(\epsilon_{\rm A}, \epsilon_{\rm B}) + F(-\epsilon_{\rm A}, -\epsilon_{\rm B})$$
(9)

$$\dot{\epsilon}_{\rm B} = -G(\epsilon_{\rm A}, \epsilon_{\rm B}) + G(-\epsilon_{\rm A}, -\epsilon_{\rm B}) \tag{10}$$

where F and G are positive functions

$$F(\epsilon_{A}, \epsilon_{B}) = \frac{K_{A}N_{A}}{v} \left\{ \left(\frac{1}{2} + \epsilon_{A}\right)^{2} p(\epsilon_{A}, \epsilon_{B}) \right. \\ \left. \times \exp\left[-\frac{a_{A}N_{A}^{2}}{v^{2}} \left(\frac{1}{2} + \epsilon_{A}\right)^{2} p^{2}(\epsilon_{A}, \epsilon_{B}) \right] \right\} (11)$$
$$G(\epsilon_{A}, \epsilon_{B}) = \frac{K_{B}N_{B}}{v} \left\{ \left(\frac{1}{2} + \epsilon_{B}\right)^{2} q(\epsilon_{A}, \epsilon_{B}) \right. \\ \left. \times \exp\left[-\frac{a_{B}N_{B}^{2}}{v^{2}} \left(\frac{1}{2} + \epsilon_{B}\right)^{2} q^{2}(\epsilon_{A}, \epsilon_{B}) \right] \right\} (12)$$

and $p(\epsilon_A, \epsilon_B)$ and $q(\epsilon_A, \epsilon_B)$ are function derived from $p(\chi_L)$ and $q(\chi_L)$ respectively; here we use the same notations for convenience. To investigate the behavior near the $(\epsilon_A, \epsilon_B) =$ (0, 0) fixed point, the above equations are expanded for small ϵ 's,

$$\dot{\epsilon}_{A} = -2\left(\epsilon_{A}F_{A} + \epsilon_{B}F_{B} + \frac{1}{2}\epsilon_{A}\epsilon_{B}^{2}F_{ABB} + \frac{1}{2}\epsilon_{A}^{2}\epsilon_{B}F_{AAB} + \frac{\epsilon_{A}^{3}}{6}F_{AAA} + \frac{\epsilon_{B}^{3}}{6}F_{BBB} + \cdots\right)$$

$$\dot{\epsilon}_{B} = -2\left(\epsilon_{A}G_{A} + \epsilon_{B}G_{B} + \frac{1}{2}\epsilon_{A}\epsilon_{B}^{2}G_{ABB} + \frac{1}{2}\epsilon_{A}^{2}\epsilon_{B}G_{AAB} + \frac{\epsilon_{A}^{3}}{6}G_{AAA} + \frac{\epsilon_{B}^{3}}{6}G_{BBB} + \cdots\right)$$
(13)

where we use the notation

$$F_{A} \equiv \frac{\partial F}{\partial \epsilon_{A}} \bigg|_{(0,0)},$$
$$F_{AAB} \equiv \frac{\partial^{3} F}{\partial^{2} \epsilon_{A} \partial \epsilon_{B}} \bigg|_{(0,0)}$$

etc. to represent the corresponding derivatives and similarly for G. It is clear from eq. (13) that eigenvalues of the Jacobian matrix are $-(F_A + G_B) \pm \sqrt{(F_A + G_B)^2 - 4(F_A G_B - F_B G_A)}$, which depend on the values of the parameter v. As observed in experiments and numerical solutions of eq. (8), when v is lowered, Hopf bifurcation occurs in which the stable fixed point gives way to oscillatory limit cycle at v_c . Thus the phase boundary of the Hopf bifurcation can be solved from $(F_A + G_B)|_{v_c} = 0$ which leads to the phase diagram of v_c versus χ_0 . The emerging frequency at the Hopf bifurcation point is given by $\omega_c = 2\sqrt{(F_A G_B - F_B G_A)|_{v_c}}$. For a given value of $\chi_0 =$ $N_{\rm A}/N_{\rm B}$, the corresponding $v_{\rm c}$ is calculated from $(F_{\rm A} +$ $G_{\rm B}|_{v_{\rm c}} = 0.$ Using $p(\chi) = \{2 - [1/(1 + \chi)]\}/2$, the phase boundary for the Hopf bifurcation is shown in Fig. 5 together with the experimental data for comparison. The agreement is reasonably well though large deviation occurs for small χ presumably due to the approximation of $p(\chi) =$ $\{2 - [1/(1 + \chi)]\}/2$ is less satisfactory in this regime. To justify more rigorously the emergence of oscillation is through a supercritical Hopf bifurcation, one can apply a theorem in Guckenheimer and Holmes²⁹⁾ to the system described in eq. (13) which translates in the present case to



Fig. 5. (Color online) Phase boundary of normalized critical vibration strength vs χ_0 . Dashed curve is the theoretical result obtained from the flux model. The experimental data (symbols) are also shown for comparison.

the condition that supercritical Hopf bifurcation occurs if $(F_{AAA} + G_{AAB})F_B > (G_{BBB} + F_{ABB})G_A$. Using the function form of $p(\chi) = \{2 - [1/(1 + \chi)]\}/2$, and by explicitly calculating the derivatives, we have verified indeed the above condition holds; confirming the supercritical Hopf bifurcation. For supercritical Hopf bifurcation, the amplitude of oscillation increases continuously as $\Delta \sim (v_c - v)^{1/2}$. This is also verified by our numerical solutions of the differential equations (8). It should be noted that the possibility of a Hopf bifurcation leading to a limit cycle dynamics was also speculated by Evesque.³⁰

3.5 Saddle-node bifurcation at v_f

We have found experimentally²²⁾ that for $v < v_c$, granular clock appears and the period of the oscillation (τ) increases as v is lowered until there is no more oscillation at $v = v_f$ and the system is in a segregated/clustering state. In fact the period τ decreases with $(v - v_f)$. If one considers increasing v from a very low value, then at v_f oscillation emerges suddenly, which is a characteristic of the saddle-node (or "blue-sky") bifurcation. In fact, our numerical solution of the dynamical equations (8) indicates that new asymmetric fixed point pairs emerge for $v < v_f$. Although we have no rigorous proof that the system undergoes a saddle-node bifurcation, the numerical behavior of the systems strongly suggested it is the case.

Assuming saddle-node bifurcation occurs at v_f , one can proceed to compute the phase boundary of v_f . The calculation is somewhat tedious and we shall just outline the procedure. Defining $n_A \equiv N_{AL}/N_A$ and similarly for n_B and denoting the new asymmetric fixed point by $(n_A^*, n_B^*) \neq (1/2, 1/2)$. (n_A^*, n_B^*) satisfies

$$F(n_{\rm A}^*, n_{\rm B}^*) = F(1 - n_{\rm A}^*, 1 - n_{\rm B}^*);$$

$$G(n_{\rm A}^*, n_{\rm B}^*) = G(1 - n_{\rm A}^*, 1 - n_{\rm B}^*)$$
(14)

where F and G are positive functions

$$F(n_{\rm A}, n_{\rm B}) = \frac{K_{\rm A}N_{\rm A}}{v} \left\{ n_{\rm A}^2 p(n_{\rm A}, n_{\rm B}) \times \exp\left[-\frac{a_{\rm A}N_{\rm A}}{v^2} n_{\rm A}^2 p^2(n_{\rm A}, n_{\rm B}) \right] \right\}$$
(15)

$$G(n_{\rm A}, n_{\rm B}) = \frac{K_{\rm B}N_{\rm B}}{v} \left\{ n_{\rm B}^2 q(n_{\rm A}, n_{\rm B}) \times \exp\left[-\frac{a_{\rm B}N_{\rm B}}{v^2} n_{\rm B}^2 q^2(n_{\rm A}, n_{\rm B}) \right] \right\}.$$
 (16)

At $v_{\rm f}$, saddle-node bifurcation occurs, there is a zero eigenvalue for the Jacobian matrix²⁹⁾ for the dynamics near $(n_{\rm A}^*, n_{\rm B}^*)$, which leads to the equation for determining the phase boundary of $v_{\rm f}$: $(F_{\rm A}^*G_{\rm B}^* - F_{\rm B}^*G_{\rm A}^*)|_{v_{\rm f}} = 0$, where we use the notation

$$F_{\rm A}^* \equiv \frac{\partial F}{\partial n_{\rm A}} \bigg|_{(n_{\rm A}^*, n_{\rm B}^*)}$$

etc. Detail results and comparison with experimental data will be presented in our future work.²⁸⁾

4. Other Interesting Scenarios

The mechanism of the clustering phenomenon in monodisperse grains in a 2-compartment vibrating chamber originates from the bifurcation of a single nonlinear dynamic equation, whereas limit cycles occur for the bi-disperse grains in the case of a 2-compartment system resulted from the Hopf bifurcation of a system of two nonlinear differential equations. Thus one may speculate that for more complicated systems governed by three or more nonlinear coupled differential equations, the resulting dynamics will be richer; and can even be chaotic. Here we shall investigate several case of grains in compartmentalized systems governed by three or more coupled equations and examine the possible resulting dynamics.

4.1 Monodisperse grains in M-compartments

Consider a single type of grains in a container with M cyclic compartments and grains in each compartment can transport to two neighboring compartments through slits (see Fig. 6). This model for $M \ge 3$ is first studied refs. 13 and 16. Denote the number of grains in the *j*th compartment by N_j , using a similar flux model, the equations of motion are

$$\frac{1}{K}\dot{N}_{j} = -\frac{2N_{j}^{2}}{v}\exp\left[-a\left(\frac{N_{j}}{v}\right)^{2}\right] + \sum_{i=j\pm 1}\frac{N_{i}^{2}}{v}\exp\left[-a\left(\frac{N_{i}}{v}\right)^{2}\right]$$
(17)

where *K* has a similar definition as in previous sections, and the periodic boundary condition is understood (i.e., the index j = 0 and M + 1 is taken to be j = M and 1 respectively). It is easy to see that the equipartition solution, $N_j = N/M$ for j = 1, 2, ..., M is always a fixed point. To investigate the



Fig. 6. A M-compartment system with periodic boundary condition.

stability of this fixed point, consider $\epsilon_j \equiv N_j/N - 1/M$ and the equations become

$$\dot{\epsilon}_j = -2F(\epsilon_j) + F(\epsilon_{j+1}) + F(\epsilon_{j-1})$$
 for $j = 1, 2, \dots, M$ (18)

where

$$F(\epsilon) = \frac{KN}{v} \left\{ \left(\frac{1}{M} + \epsilon\right)^2 \exp\left[-\frac{aN}{v^2} \left(\frac{1}{M} + \epsilon\right)^2\right] \right\}$$
(19)

again the periodic boundary conditions for the index j is understood. The fixed points with the associated bifurcations and the coarsening kinetics have been studied in detail in refs. 31 and 32. No oscillation nor chaotic dynamics is reported.

4.2 Bi-disperse grains in M-compartments

Consider now two types of grains A and B in a container with M cyclic compartments, using the flux model, the governing dynamical equations are

$$\frac{1}{K_{A}}\dot{N}_{Aj} = -\frac{2N_{Aj}^{2}}{v_{A}(\chi_{j})}\exp\left[-a_{A}\frac{N_{Aj}^{2}}{v_{A}^{2}(\chi_{j})}\right] \\ + \sum_{k=j\pm 1}\frac{N_{Ak}^{2}}{v_{A}(\chi_{k})}\exp\left[-a_{A}\frac{N_{Ak}^{2}}{v_{A}^{2}(\chi_{k})}\right] \\ \frac{1}{K_{B}}\dot{N}_{Bj} = -\frac{2N_{Bj}^{2}}{v_{B}(\chi_{j})}\exp\left[-a_{B}\frac{N_{Bj}^{2}}{v_{B}^{2}(\chi_{j})}\right] \\ + \sum_{k=j\pm 1}\frac{N_{Bk}^{2}}{v_{B}(\chi_{k})}\exp\left[-a_{B}\frac{N_{Bk}^{2}}{v_{B}^{2}(\chi_{k})}\right] \\ \text{for } j = 1, 2, \dots, M$$
(20)

where $\chi_k \equiv N_{Ak}/N_{Bk}$ is the ratio of A to B grains in the *k*th compartment, and v_A and v_B have similar definitions as in the case of 2-compartments. Again the periodic boundary conditions for the index *j* is understood for cyclic compartments. Note that the conditions $\sum_{k=1}^{M} N_{Ak} = N_A$ and $\sum_{k=1}^{M} N_{Bk} = N_B$ hold and the actual number of independent coupled equations is 2(M - 1). The simplest case for cyclic compartment is for M = 3 (with 4 coupled equations) and some preliminary results were presented in ref. 33 in which some irregular or random dynamical behavior was reported suggesting the possibility of chaos. On the other hand, if the compartments are arranged linearly (non-cyclic) with walls on both ends, regular periodic oscillations were observed in ref. 33.

4.3 Tri-disperse grains in 2-compartments

This is a scenario resulting in only three nonlinear coupled dynamical equations and could possibly be the simplest case that would show chaotic dynamics. Consider three types of grains A, B, and C in a system consisting of left and right compartments. Denoting the ratios of grains pairs by $\chi_B \equiv N_A/N_B$ and $\chi_C \equiv N_A/N_C$. Using similar method as in previous sections, one arrives at the governing dynamical equations

$$\frac{1}{K_{\rm A}}\dot{N}_{\rm AL} = -\frac{N_{\rm AL}^2}{v_{\rm AL}}\exp\left(-a_{\rm A}\frac{N_{\rm AL}^2}{v_{\rm AL}^2}\right) + \frac{(N_{\rm A} - N_{\rm AL})^2}{v_{\rm AR}^2}\exp\left[-a_{\rm A}\frac{(N_{\rm A} - N_{\rm AL})^2}{v_{\rm AR}^2}\right]$$

$$\frac{1}{K_{\rm B}}\dot{N}_{\rm BL} = -\frac{N_{\rm BL}^2}{v_{\rm BL}}\exp\left(-a_{\rm B}\frac{N_{\rm BL}^2}{v_{\rm BL}^2}\right) +\frac{(N_{\rm B} - N_{\rm BL})^2}{v_{\rm BR}}\exp\left[-a_{\rm B}\frac{(N_{\rm B} - N_{\rm BL})^2}{v_{\rm BR}^2}\right) \frac{1}{K_{\rm C}}\dot{N}_{\rm CL} = -\frac{N_{\rm CL}^2}{v_{\rm CL}}\exp\left(-a_{\rm C}\frac{N_{\rm CL}^2}{v_{\rm CL}^2}\right) +\frac{(N_{\rm C} - N_{\rm CL})^2}{v_{\rm CR}}\exp\left[-a_{\rm C}\frac{(N_{\rm C} - N_{\rm CL})^2}{v_{\rm CR}^2}\right]$$
(21)

where $a_{\rm C}$ is defined in a similar way as $a_{\rm A}$ in previous section, $v_{\rm AL} \equiv v/p_{\rm A}(\chi_{\rm BL}, \chi_{\rm CL})$ and similarly for $v_{\rm BL}$, $v_{\rm CL}$, etc. The functions $p_{\rm A}$, $p_{\rm B}$, and $p_{\rm C}$ have similar roles as the functions p and q for the bidisperse case in previous sections. Note that $N_{\rm B}/N_{\rm C} = \chi_{\rm C}/\chi_{\rm B}$ and hence it is sufficient to use $\chi_{\rm B}$ and $\chi_{\rm C}$ as independent variables for the functions $p_{\rm A}$, $p_{\rm B}$, and $p_{\rm C}$. The detail dynamics of this system will be reported elsewhere.

5. Concluding Remarks

In this article, we have explained the notions of unstable evaporation/condensation and multiple temperatures for a heterogeneous granular gas system. These are unique properties of granular gases and there is no counter part of these concepts for a molecular gas in thermal equilibrium. With these notions, we can understand the intriguing phenomena of Maxwell's demon as well as the granular clock. These phenomena are possible because of the nonequilibrium nature of the system and the strong nonlinear dependence of local energy dissipation on the configuration of the system (granular density distribution); giving rise to a wide varieties of interesting stationary as well as dynamical patterns. However, our studies are being carried out in a self consistent manner by assuming the existence of multiple granular temperatures in a heterogeneous system. It would be important to establish the existence of these multiple granular temperatures by more detailed experiments. For example, measurement of fluxes in experiments would provide independent tests of our assumption. Experiments of this kind are now well in progress and their results will be reported soon.

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