The origin and nature of Boson Peak —the normal mode analysis of disordered granular crystals

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Despite extensive theoretical [1,6] and experimental studies [7,12], a longstanding puzzle in condensed matter physics remains regarding the origin and nature of “Boson peak” (BP), where the vibrational density of states (DOS) in glasses possesses an excess of states compared with the crystalline counterpart. Here we show that BP is successfully observed in 2D hexagonal granular packing, where the disorder is due to the force network, i.e. the spatial heterogeneity of elasticity [6]. Using photo-elastic techniques [13], the disordered particle interaction can be precisely measured to resolve the origin and nature of BP for the first time in a real scenario. We discover the structures of DOS of disordered crystals reminiscent of the corresponding perfect crystals, which is consistent with the recent studies in SiO₂ [8] and in disordered gels [14], notwithstanding the drastically different systems. Moreover, we propose a mechanism to clarify that BP is not merely a broadened and shifted van Hove singularity [2,7] but instead it is due to an interplay of the mesoscopic screening effect and the microscopic elasticity disorder – causing, respectively, the broadening and the attenuation of the first and the second van Hove singularity. This may lead to an in-depth understanding of BP in structure glasses.

Keywords:

Introduction Normal-mode analysis serves a powerful tool in understanding the vibrational properties of crystalline solids, where each mode has a well defined physical meaning as plane-wave due to the translational symmetry [15,16]. In disordered and amorphous systems, such as structure glasses and disordered crystalline solids with heterogeneous interactions, the nature of the normal modes is still open to debate [17]. In particular, the origin and nature of BP have become the focus because of its fundamental importance in understanding the anomalous behaviors of thermodynamical variables of these systems. A series of theoretical models have been proposed such as the mode coupling model [18], the potential energy landscape model [19], the broadening of the van Hove singularity [2,12], the spatial heterogeneity of elastic moduli [3,6], the jamming near isostaticity [20,21], and the crossover of transverse phonon at the Ioffe-Regel limit [4], et al.; at the same time, a wide range of experiments have been performed in drastically different systems, from macroscopic probes in molecular glasses using neutron and X-ray scattering [7,8] to microscopic probes in colloidal glasses of structure [10,12] or interaction disorders [9] and in vibrated granular media [22]. Despite tremendous work, the origin and nature of BP remain a deep and serious mystery in physics, even in crystals with disordered interactions, which is the main focus of this work.

In this study, we tackle this challenging problem by directly measuring the dynamical matrix of disordered granular crystals. BP is clearly observed: as pressure increases, its height decreases and its frequency increases; moreover, a novel shoulder structure gradually develops in the derived DOS, D(ω)/ω, and, correspondingly, a second peak gradually emerges in the DOS, D(ω), where the two peaks show a good alignment with the two van Hove singularities of the corresponding perfect hexagonal packing. This provides a strong experimental evidence that BP is not only related to the broadening of the first van Hove singularity but also related to the attenuation and eventual disappearance of the second van Hove singularity. Moreover, because of the spatial heterogeneity of elastic moduli, the second peak of D(ω) and hence the second singularity are strongly affected due to the nearest neighbour interaction (NNI) whereas the BP and hence the first singularity are weakly affected due to the second nearest neighbour interaction (SNNI) – creating an effective screening to shield local elastic modulus fluctuations, as directly measured in the experiment.

Results and discussion For each packing, the dynamical matrix M was constructed once the stiffness constants were readily obtained from M. Figure 1(a) shows D(ω) at different pressure P: when P ≤ 3.15N/m, there is only a single peak followed with a knee structure that gradually transforms to the second peak when P > 3.15N/m. Correspondingly in

![FIG. 1: (a) The density of states D(ω) as a function of angular frequency ω at six different pressure. (b) The derived density of states, D/ω, as a function of ω at six different pressure. These curves are obtained by normalizing D(ω) with the Debye Scalling – linear in ω in 2D. Each curve in (a,b) is ensemble averaged over five different runs at the same pressure.](image-url)
The probability distribution (PDF) of the spring constant is shown in Fig. 1b. BP is followed by a knee structure, nearly invisible at $P=2.06$ N/m. In contrast, only a single peak was observed in the previous colloidal experiments [9].

In literature, a key observation is that the spatial distribution of $|\omega|$ changes its characteristics across BP [9,12]. When $\omega < \omega_0$, it is more coherent in space with cooperative variations of the vector orientation and magnitude: in disordered colloidal crystals, it is plane-wave like [9]; whereas in colloidal structure glasses, it is either plane-wave like [11] or a superposition of a plane-wave-like background and some quasi-localized structures [10]. Conversely, when $\omega \approx \omega_0$, it becomes more random in space [9,12]. Our results are qualitatively similar to Refs. [9,12], as shown in Fig. 2 which suggests the universality of such behavior and a negligible role of the thermal fluctuation.

To clarify the possible connection between the two peaks in Fig. 1b and the two van Hove singularities of the hexagonal packing, we compute $D(\omega)$ of the perfect hexagonal packing by setting individual $k$ equal to the mean value $\langle k \rangle$ of the disordered crystal. Figure 3a shows $D(\omega)$ of both the disordered granular crystal (open circles) and the perfect granular crystal (solid circles) for direct comparison, where they show an excellent alignment, consistent with the recent findings in the SiO$_2$ experiment [8] and in the numerical study of disordered gel models [14] in spite of the marked differences of these systems. As shown in SI, $\omega_0$, $\omega_0$, and $\omega_0$ of the first van Hove singularity have a one-to-one correspondence. Nevertheless, some important questions emerge, in particular: why the second peak of $D(\omega)$, hence the second singularity, vanishes eventually; why the first van Hove singularity evolves to the Boson peak and not the second one, et al.

In order to address the above questions, we first investigate the two van Hove singularities as the starting point, which are related to the extreme points of the dispersion relation [15,16] (See Fig. 9 of SI). The first singularity corresponds to the transverse waves of the $\Sigma$ lines with wave vector $q$ reaching the six $M$ points of the first Brillouin zone, corresponding to the six red dashed lines in real space as depicted in Fig. 3b with $q_{\Sigma} = \sqrt{3}d$ and $d$ being the disk diameter. The second singularity corresponds to the longitudinal waves of the $T$ lines with $q_{\Sigma} = 1.85d$, which are equivalent to the six green solid lines in real space as depicted in Fig. 3b. (See...
the early discoveries that the acoustic wave changes its nature as the strong link\[24\]. Second, we have confirmed with significant differences of these systems – suggesting the disordered feature of glassy materials. Here coarse-grain is applied using a Gaussian function of standard deviation $\frac{1}{2}d$, as described in detail in Ref.[23]. In Fig. 4(a), the $k$ of NNI fluctuates more rapidly in space with many small-scale structures. In contrast, the variation of $k$ of SNNI is much more smoother in space as shown in Fig. 4(b). Since $k$ of SNNI is essentially shear modulus, its spatial heterogeneity in Fig. 4(b) is consistent with the findings of Ref.[6].

Conclusions By studying the disordered granular crystals, we have made several important discoveries regarding the origin and nature of Boson peak. First, Boson peak is deeply connected with the first van Hove singularity and they have a one-to-one correspondence when comparing disordered and perfect crystals of the same mean stiffness constant, which is consistent with the early proposition and findings [1][2][7] and, in addition, is also consistent with the recent findings in SiO$_2$ glasses [8] and in disordered gels [14] despite of the significant differences of these systems – suggesting the disorder as the strong link [24]. Second, we have confirmed with the early discoveries that the acoustic wave changes its nature across the Boson peak [9][12]. Lastly, and not the least importantly, we clarify how disorder plays its role in transforming the first van Hove singularity into a Boson peak at different scales: at the microscopic scale, disorder causes the significant attenuation and the eventual vanishing of the second van Hove singularity; whereas at the mesoscopic scale, disorder is mostly screened by the effective cluster-cluster interaction; moreover, at even larger scales, disorder is restored due to the spatial heterogeneity of elastic modulus. We believe a similar type of mechanism may be extended to understand Boson peak in structural glasses, which will be our future goal.

Methods

Experimental methods. A densely packed granular crystal is naturally disordered because of the internal force network structure [25]. To create packing of different pressure, we used a “biax” similar to the one reported in [26][27]. Different from the “biax” in [26][27], here four walls can move symmetrically while the center of mass is fixed. As shown in Fig. 3(a) of SI, the “biax” consists of a rectangular frame filled with ~1500 monodisperse disks (diameter 1.4 cm) manually deposited on a powder-lubricated glass surface to construct a hexagonal packing. Starting from a square, biaxial compression is applied in order to create packing of a given pressure. In a given experiment, ten runs were performed following the same protocol to create an ensemble. In each run, two images of the photoelastic disks were taken in order to record the position and stress of the disks [27]. Two sample images are shown in Fig. 5(b-c) of SI. Applying image processing and force-inverse algorithm allows us to determine the geometry of each particle, the contacts and the contact forces between particles [13][26][27]. Using a pre-calibrated curve of contact force versus deformation, we can determine the stiffness constant of each contact (Details in SI). Figure 5(d) of SI shows a reconstructed elastic network, where the packing is mapped onto a lattice with point mass at each node connected with prestressed springs with the line thickness proportional to the stiffness.

Friction between disks. In this experiment, disks are essentially frictionless due to the following reasons: (1) the hexagonal packing structure has six contacts per particle, which is the maximum value for a 2D packing; (2) each disk has been wrapped around its cylindrical surface with a teflon tape of a static friction coefficient of 0.1. Hence, we estimate that the effective frictional force component is two orders of magnitude smaller compared with the normal force component at a given contact. Therefore, the disks are essentially frictionless.

Normal mode analysis. In order to apply the normal mode analysis, we first construct the dynamical matrix $M$ of the system. The system is modeled as an elastic triangular lattice of $N$ nodes of mass $m$, connected by $ZN$ springs of equilibrium spring length, to a very good approximation, equal to the disk diameter. For the Hexagonal packing of the granular crystal, the coordination number $Z = 6$. Following a similar method described in Ref.[21][29], we can take a Taylor expansion of the elastic energy of the system: 

\[
V(Z) = \frac{1}{2} \sum_{NN} k_{NN} (Z - Z_{0})^2 + \frac{1}{4} \sum_{NNNN} k_{NNNN} (Z - Z_{0})^4 + \cdots
\]

where $k_{NN}$ and $k_{NNNN}$ are the stiffness constants of the nearest and second nearest neighbour interactions, respectively, and $Z$ is the coordination number of each node. The dynamical matrix $M$ is given by 

\[
M = \sum_{NN} k_{NN} \mathbf{e} \mathbf{e}^T + \sum_{NNNN} k_{NNNN} \mathbf{e} \mathbf{e}^T \mathbf{e} \mathbf{e}^T
\]

where $\mathbf{e}$ is the unit vector along the direction of the force. The eigenvalues of $M$ are the squared frequencies of the normal modes of the system and the eigenvectors are the displacement fields of these modes. The normal mode analysis allows us to determine the relationship between the stiffness and the equilibrium structure of the system.
around its equilibrium position for an arbitrary infinitesimal displacement field $\dd{u}$. This will yield the quadratic energy term $\delta E = \frac{1}{2} \sum_{\langle i,j \rangle} k_{ij}(u_{ij}^2 - f_{ij} |u_{ij}|)$, where the summation is over all springs and the indices $(i,j)$ stand for the spring connecting from particle $i$ to particle $j$. Here $u_{ij}$ and $u_{ij}$ are respectively the parallel and perpendicular components of the relative displacement between particles $i$ and $j$ projected onto the line connecting the centers, i.e. the lattice points of these two particles; $k_{ij}$ is the stiffness of the spring; $f_{ij}$ is the magnitude of the contact force between the particle $i$ and the particle $j$; $r_{ij}$ is the constant equilibrium spring length – equal to the disk diameter. The dynamical matrix $M$ can then be obtained from the expression of $\delta E$ by expressing the terms of $u_{ij}$ and $u_{ij}$ using the $x$ and $y$ components of displacement field $\dd{u}$ at the lattice points of particle $i$ and particle $j$ (see e.g. Refs. [21, 29] for detail). The dynamical matrix $M$ can be diagonalized to find its eigenvalues $\lambda_n = \frac{1}{2} m \omega_n^2$ and the corresponding eigenvectors $|\alpha_n\rangle$ which can provide the distribution of the polarization vector in space.

The effective spring constant between the second nearest neighbour particles. Approximations are made in order to measure the effective stiffness $k_{\text{eff}}$ between two second nearest neighbor disks, e.g. the disks $i$ and $j$ in Fig. 3(c). We assume that $k_{\text{eff}}$ is predominantly due to their nearest neighbour particles, i.e. those green particles in Fig. 3(c). Here only the minimum core of cluster particles are considered; in reality the inclusion of more particles will only consolidate our argument presented in the main text. From the elasticity theory, $k_{\text{eff}} = \frac{E}{\sqrt{3}}$, where $E$ is the Young’s modulus and $\sqrt{3}$ is a geometrical factor. To compute $E$, we apply $E = 9BG/(3B + G)$ by treating the cluster as a 2D cylinder [28], where $B$ and $G$ are respectively the bulk and shear modulus of the cluster. To measure $B$ and $G$, we use the method described in detail in Ref. [29], which applies a small compression (or shear) strain $\gamma$ to every disk of the cluster including the freely deformable boundary disks of the cluster as painted in blue in Fig. 3(c). This allows us to compute the stress tensor $\sigma_{ij}$ of the cluster to find $B$ and $G$ using $B = \frac{\sigma_{yy} + \sigma_{xx}}{2\gamma}$ and $G = \frac{\sigma_{xx} - \sigma_{yy}}{2\gamma}$. Finally, we can determine $k_{\text{eff}}$. Note that in Fig. 3(d-e), the subscript $\text{eff}$ is dropped.

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The corresponding spring network. Here the thickness of each in-
sheets in order to record the stress fluctuations inside the packing.(d)
cause of the existence of the self-organized force network in-
side the system, which can be directly observed with a pair
of circular polarizers in place as shown in Fig. 5(c). Apply-
ing a biaxial setup sketched in Fig. 5(a), which mainly con-
nects with a force gauge (IMADA DS2-5N with a resolu-
tion of 0.01N). A disk was initially placed within the rectan-
gular box. When the disk was barely touched by the two walls,
the force gauge was set to zero. The compression was grad-
dually applied with positions closely monitored using a high-
precision linear translation stage with a 1

\[ P \times 0.01 \text{N} \]

\[ \langle k \rangle \]

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\[ 0.01 \text{N} \]

to find

\[ k = \frac{df_n(\delta r)}{d\delta r} \]

The stiffness disorder \( \delta k/\langle k \rangle \). The ratio \( \delta k/\langle k \rangle \) provides a quantitative characterization of the disorder of the stiffness constant \( k \) in space. Here \( \delta k \) is the standard deviation and \( \langle k \rangle \) is the mean. Figure 7 shows \( \delta k/\langle k \rangle \) as a function of pressure \( P \). Within the pressure range in the experiment, \( \delta k/\langle k \rangle \) decreases approximately linearly with \( P \), albeit with some fluctuations of magnitude \( \sim 0.01 \).

The relationship between \( \omega_b \), \( \omega_p \), and \( \omega_c \). As discussed in detail in the main text, the first peak frequency of \( D(\omega) \) is denoted as \( \omega_p \). Similarly, the first peak frequency of \( D(\omega)/\omega \) is the Boson peak frequency \( \omega_b \). For \( D(\omega) \) of the perfect crystal with all \( k \) equal to \( \langle k \rangle \) of the disordered crystal at the same pressure, the frequency of the first Van Hove singularity is denoted as \( \omega_c \). Typically \( \omega_b < \omega_p < \omega_c \). However, there is a one-to-one correspondence between these three frequencies as seen in Fig. 5 where different data points of panels(a-c)

**FIG. 6:** The calibrated curve of contact force \( f_n \) versus deformation \( \delta r \). The data points have been ensemble averaged by repeating the calibration 10 times for each disk and for five randomly chosen disks. The blue solid line is a power law fit of the expression: \( f_n = 3.09 \times 10^{6} \times \delta r^{1.89} \).

**FIG. 5:** (a) A schematic of the experimental setup. The setup is a biax with a square frame filled with \( \sim 1500 \) photo-elastic disks. For a given granular packing, isotropic compression can be applied simultaneously moving the four walls inwards. (b) A small portion of the hexagonal packing of a granular crystal, where the disk diameter is 1.4 cm. This image was taken without polarizer sheet in order to record the particle configurations. (c) The corresponding stress image of the disk packing in (b). This image was taken with polarizer sheets in order to record the stress fluctuations inside the packing.(d) The corresponding spring network. Here the thickness of each individual line segment is proportional to the magnitude of the spring constant of every contact between neighbouring disks as shown in image (c).

**Supplementary Information**

**Experimental setup.** The experiment was performed using a biaxial setup sketched in Fig. 5(a), which mainly consists of a rectangular frame filled with \( \sim 1500 \) mono-disperse photo-elastic disks with a size of 1.4 cm in diameter. These disks are densely packed into a crystal of hexagonal packing as shown in Fig. 5(b). Starting from a square, isotropic compression was applied by simultaneously moving the four walls inward to deform the system to a smaller square while keeping the center-of-mass position fixed. Depending on the amount of strain applied, the packing of desired pressure can be prepared. Each hexagonal packing forms a disordered crystal because of the existence of the self-organized force network inside the system, which can be directly observed with a pair of circular polarizers in place as shown in Fig. 5(c). Applying image processing and data analysis can transform a stress image in Fig. 5(c) to a spring network in Fig. 5(d), as will be discussed in the next section.

**The calibration of the \( f_n(\delta r) \) curve.** In order to obtain the stiffness constant \( k \) at a given contact between two particles, we need to calibrate the relationship between the normal force \( f_n \) and the deformation \( \delta r \) of the contact. The calibra-

\[ f_n(\delta r) = k \delta r \]

\[ k \delta r \]

\[ f_n \]

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\[ \delta r \]

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represent results of different pressure $P$. The one-to-one relationship can be quantified using the linear fit in each panel of Fig. 8(a-c). Moreover, $\omega_p$ increases nonlinearly with $P$ and can be fit with $\omega_p = 600.6 \times P^{0.30}$ as shown in Fig. 8(d).

The dispersion relation of the perfect hexagonal packing. For a perfect hexagonal packing of constant stiffness, the dispersion relation $\omega(k)$ can be found analytically. Figure 8 plots $\omega(k)$ with wave vector $k$ varying along the $\Sigma$ line, the $M_L$ line, and the $T$ line in the first Brillouin zone – a hexagon in reciprocal space as drawn in the inset. The $\Sigma$ lines represent the six high-symmetry lines connecting the center of the Brillouin zone, i.e. the point $\Gamma$, to the mid-points of the six lat- erals. Similarly, the $T$ lines represent the six high-symmetry lines connecting the center point $\Gamma$ to the six vertices of the Brillouin zone. In the inset of Fig. 9 we only plot one $\Sigma$ line and $T$ line, along with the line connecting the mid-point $M$ and the vertex $L$. These three lines form a rectangular trian-gle. Due to the symmetry of the Brillouin zone, the dispersion relations of other triangles are identical. Note that only along the high symmetry lines such as the $\Sigma$ lines and $T$ lines, a plane wave can be decomposed into a transverse or a longitudi-nal wave, e.g. along the line $\Gamma M$ and the line $\Gamma L$. Figure 9 shows that there are only three points satisfying $\frac{d \omega}{d k} = 0$: the point $\omega_1$ (transverse waves on $\Sigma$ lines) corresponds to the first van Hove singularity on the density of states $D(\omega)$; the point $\omega_2$ (longitudinal waves on $\Sigma$ lines) corresponds to the max-i-mum frequency of $D(\omega)$; the point $\omega_3$ (longitudinal waves on $T$ lines) corresponds to the second van Hove singularity of $D(\omega)$. Note that $\omega_2 \approx 1.60 \omega_1$ and $\omega_3 = \sqrt{3} \omega_1$ and these three frequencies match exactly to the values obtained numerically from the hexagonal packing at the same pressure.

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Additional Information
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