Amplitude-dependent phononic processes in a diatomic granular chain in the weakly nonlinear regime

Jérémy Cabaret, Vincent Tournat, and Philippe Béquin
LAUM, CNRS, Université du Maine, Avenue O. Messiaen, 72085 Le Mans, France
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Nonlinear acoustic processes of second harmonic generation and nonlinear resonances in a diatomic granular chain (a granular phononic crystal) with static precompression are reported. The observed nonlinear self-action process which manifests itself as shifts in resonance frequencies of the chain leads to amplitude-dependent band edges: the properties of the phononic crystal change as a function of wave amplitude. Observed nonlinear effects at the band edges are exceptionally strong (self-induced attenuation and self-induced transparency) due to the peculiar frequency dependence of the attenuation in these frequency regions. The reported effects open the way for applications in wave tailoring by nonlinear phononic crystals, using amplitude-dependent processes, such as passive amplitude-dependent attenuators or amplifiers and various logical elements.

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I. INTRODUCTION

Wave propagation in granular chains, composed most of the time of spherical particles arranged on a line, has been the subject of numerous theoretical, experimental, and numerical studies in the last few decades [1–12]. Primary applications of these apparently simple media were devoted to the attenuation of strong mechanical shocks, arising for instance from explosions [3,4,13]. More recently, these media have been used as model structures for the study of fundamental wave processes such as soliton propagation or interaction, splitting or mitigation, interaction with a boundary, defect modes, discrete breathers, and intrinsic localized modes [14–21]. Also, new applications have recently been foreseen for elastic wave control: filtering devices, rectifiers, actuating devices, and sound scramblers [15], to cite some. The two main ingredients that place these granular chains at the origin of many potential applications are their wave dispersion properties and their nonlinear character [22]. The possible strong frequency dependence arises from the periodicity of the structure (eventually disturbed a bit by a defect or a property gradient), which gives rise to phononic effects, among which are forbidden band gaps. Nonlinearities are caused by the mechanical behavior of the contacts between elastic beads and can reach levels much higher than those encountered in homogeneous bulk media [7,23,24]. The strong frequency dependence of the attenuation and the strong nonlinearity are both at the origin of the first proposed acoustic diode or rectifier [25,26].

Here, we report on wave-amplitude-dependent effects in a diatomic granular chain that could give rise to wave-tailoring applications for sound and ultrasound in solids. A diatomic granular chain is composed of two types of particles of different masses that are alternatively placed. It exhibits two propagation bands and two stop bands. In comparison to a monatomic granular chain, this leads to three band edges with strong changes in attenuation as a function of frequency, instead of a single one. Our interest resides in the fact that different scenarios of amplitude-dependent effects are covered, which makes this configuration consequently richer than that of a monatomic chain. We show that even in a weakly nonlinear granular chain, self-action effects may be exceptionally strong for some peculiar frequency bands, located at the band edges. Similar effects have been predicted recently using the harmonic balance method and some numerical simulations in various nonlinear phononic crystals [22]. The experimental results presented here confirm the possibility of such effects with however a different quantitative behavior because of the different type of nonlinearity involved.

First, we present the experimental setup and recall the characteristics of the linear acoustic behavior of such a precompressed diatomic granular chain. Then, the nonlinear equations of motion are derived up to quadratic order in bead displacement, by assuming a precompression level higher than the acoustic one. Solutions for the second harmonic generation are obtained and compared with experimental results. Nonlinear self-action effects, i.e., effects at the excitation frequency itself, are also reported: nonlinear resonance frequency shift and nonlinear shift of the resonance quality factor. The latter effects lead (at particular frequencies close to the band edges where fast variations of the transmission as a function of frequency occur) to dramatic amplitude-dependent attenuation or transparency.

II. EXPERIMENTAL SETUP

The one-dimensional diatomic granular crystal with a total length of 26 cm is built by alternating 20 magnetic stainless steel cubes (with an edge length of 5 mm) and 20 nonmagnetic stainless steel spheres (with a radius of 4 mm). The cubes and spheres are constituted of the same material but their volume is different (and hence their mass is different) while all contacts between them are equivalent. The experimental setup is shown in Fig. 1 and the bead parameters are collected in Table I. The first bead is in contact with a piezoelectric (CEDRAT) actuator hung from a rigid structure. The use of magnetic cubes allows us to self-sustain the medium in the air, which avoids the need for supporting walls to confine the beads as in some previous studies [9,12,16,20,21]. This eliminates

*vincent.tournat@univ-lemans.fr
FIG. 1. Schematic of the experimental setup. The weakly nonlinear diatomic granular chain is composed of steel spheres and magnetic cubes of masses \( m_1 \) and \( m_2 \), respectively. It is self-suspended, which avoids any source of friction or sound transmission within solid walls. The useful bead properties are given in Table I.

The contribution of friction from the supporting medium as well as wave transmission through it. This absence of friction is of primary importance in the observation of linear elastic wave processes but is also important for nonlinear elastic wave processes. Additionally, this produces a static precompression of \( \sim 4N \) between the beads. This precompression force is assumed to be equal to the measured pulling force necessary to disconnect two adjacent grains of the chain. Under these conditions, the effect of gravity on the contact precompression is negligible. At the other end of the chain an accelerometer (PCB) with a small mass (\( \sim 0.2 \) g) compared to the beads is attached for achieving detection. The received accelerometer signal is preamplified and finally sent to an acquisition device.

The excitation signal is a exponentially swept sine of 5 s duration with frequencies from 1 to 45 kHz. The same shape signals with increasing excitation amplitudes are successively launched in the medium. This signal is particularly well adapted to a recently developed signal processing method [27]. This nonlinear deconvolution method is able to measure the transfer functions between the excited signal and the detected one at the excited frequency but also at the harmonics of the excited frequency. In contrast, a conventional spectrum analyzer in sweep mode only provides a transfer function at the excitation frequency. Consequently, the nonlinear convolution method is used here to extract the transfer functions of the diatomic chain at the fundamental frequency (such as a spectrum analyzer in sweep mode) and at the second harmonic, with a high precision in frequency and amplitude [27,28].

### III. THEORY

#### A. Linear and weakly nonlinear equations of motion

For the theoretical description of the nonlinear wave propagation in a diatomic granular chain with precompression we consider the medium as semi-infinite, with the wave excitation at the boundary. We limit ourselves to sufficiently low frequency phenomena such that no individual vibration mode of the cubes and spheres are excited nor are there waves inside these grains. The first individual modes of the grain are estimated to occur at frequencies higher than few hundred kilohertz, while our maximum wave frequency is less than 100 kHz. In this case, the phenomena under investigation can be considered as quasistatic for the deformation of each grain, and the discrete feature of the chain is justified. Elastic deformations are concentrated near the contact zones between spheres and cubes, and the bulk material of the grains is not deformed and thus acts as a rigid mass. Therefore it is possible to model the granular chain as a chain of masses interacting by nonlinear springs with a Hertzian force-displacement law [14,29,30]. The system of \( n \) equations of motion for a monatomic granular chain is usually written as [3],

\[
m_n \ddot{u}_n = A[\delta_0 + u_{n-1} - u_n] - A[\delta_0 + u_n - u_{n+1}], \tag{1}
\]

where \( u_n \) is the displacement of the \( n \)th bead \((n \in \{1, \ldots, N\})\) around the equilibrium position, \( m_n \) is the mass of the bead \( n \), \( A = 2(R_e/2)^{1/2}(3E)/(1 - \nu^2) \) is a coefficient depending on the bead properties [with \( R_e \) an equivalent radius defined by \( R_e = (2/R_1 + 1/R_2)^{-1} \)] and \( E \) and \( \nu \) the Young’s modulus and Poisson coefficient, respectively, of the bead material, and \( \delta_0 \) is the distance of static overlap between adjacent beads. The exponent \( p = 3/2 \) is given by the Hertz potential law between adjacent particles [30]. Notice that the contact interaction between two spheres is of the same nature (the same exponent \( 3/2 \)) as between a sphere and a cube. Contact laws only differ by the proportionality constant.

In a precompressed granular chain these equations of motion can be simplified in the regime \(|u_n - u_{n-1}| \ll |\delta_0|\) [31]. The power function of Eq. (1) with \( p = 3/2 \) is expanded in a Taylor series in the form of a linear term and nonlinear quadratic, cubic, and higher-order terms in strain [10,31]. This is the so-called weakly nonlinear regime of the granular chain. Then, using the method of successive approximations, it is possible to find solutions of the system of nonlinear equations to the desired level of approximation.

By using the same procedure as in Ref. [31], the system of nonlinear equations for a diatomic granular chain composed of two types of grains with different masses can be rewritten

### Table I. Bead properties of the experimental setup.

<table>
<thead>
<tr>
<th>Cubes</th>
<th>Spheres</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cube</td>
<td>Sphere</td>
</tr>
<tr>
<td>Edge ((R_1)) or radius ((R_2)) (mm)</td>
<td>5</td>
</tr>
<tr>
<td>Density (kg/m(^3))</td>
<td>7850</td>
</tr>
<tr>
<td>Young’s modulus (GPa)</td>
<td>203</td>
</tr>
<tr>
<td>Poisson ratio</td>
<td>0.3</td>
</tr>
<tr>
<td>Mass (g)</td>
<td>1.0</td>
</tr>
</tbody>
</table>
up to the quadratic terms as

\[ m_1 \ddot{u}_n = \alpha [v_n - 2u_n + v_{n-1}] - \frac{\beta}{2} [v_n - 2u_n + v_{n-1}] [v_n - v_{n-1}], \]  

(2)

\[ m_2 \ddot{v}_n = \alpha [u_{n+1} - 2v_n + u_n] - \frac{\beta}{2} [u_{n+1} - 2v_n + u_n] [u_{n+1} - u_n], \]  

(3)

where \( \alpha = 3A \delta_0^{-2} (8R_s)^{1/2} \) is the linear stiffness of the precompressed contacts predicted by Hertz’s law [30], \( u_n \) and \( v_n \) are the displacements of the spheres and cubes, respectively, and \( \beta = \alpha R_s/(2\delta_0) \) is the parameter of quadratic nonlinearity. The \( \gamma \) symbol denotes the double derivative over time. The value \( \delta_0 \) depends on the precompression force \( F_0 \) due to the magnetic attraction force as \( \delta_0 = (F_0/A)^{2/3} \).

In order to solve this system of nonlinear equations we make use of the above-mentioned successive approximation method [31–33]. It consists in separating the equations having leading order terms (linear in displacement) from equations with second-order terms (quadratic in displacement), by considering that \( |u_n - v_n| \ll 1 \) and \( |u_n - v_{n-1}| \ll 1 \). Then, we need to solve the system of linearized equations of motion, input the solution into the quadratic term of the second-order system of equations, and solve this system of inhomogeneous linear equations.

B. Dispersion relation and linear solution

A linear solution of the problem is obtained after linearization of the system (2) and (3), i.e., omitting the quadratic in displacements terms. We consider a monochromatic wave excitation at pulsation \( \omega \). By omitting the term \( e^{i\omega t} \) the solution can be expressed as a propagating wave in the direction of increasing \( n \):

\[ u_n^l = A e^{-i\omega n}, \]  

(4a)

\[ v_n^l = B e^{-i\omega n}, \]  

(4b)

where \( k \) is the wave number and \( a = R_1/2 + R_2 \) is the distance between the centers of the sphere and the cube. After substitution of this solution into the system of linearized equations of motion, the dispersion relation is obtained:

\[ \omega_{\pm} = \sqrt{ \frac{\alpha (m_1 + m_2)}{m_1 m_2}} \pm \sqrt{ \frac{\alpha (m_1 + m_2)^2 - 2m_1 m_2 [1 - \cos (ka)]}{m_1 m_2}} \]  

(5)

This relation shows that there exist two possible frequencies \( \omega_{\pm} \) for one given \( k \), i.e., two modes of propagation. By inverting this relation, four different frequency regions are found corresponding to passbands (where the imaginary part of \( k \) is very small compared to its real part) and band gaps (where the imaginary part and the real part of \( k \) are of the same order). The two passbands are denoted as “acoustical” for the low-frequency one and “optical” for the higher-frequency one, similarly to the terms used in solid state physics [34]. Frequency limits of these bands are found for the peculiar values 0 and \( \pi/a \) of the wave number \( k \) and for \( m_1 > m_2 \):

\[ \omega_{1c} = \omega_{-} \left( k = \frac{\pi}{a} \right) = \sqrt{ \frac{2\alpha}{m_1}}, \]  

(6)

\[ \omega_{2c} = \omega_{+} \left( k = \frac{\pi}{a} \right) = \sqrt{ \frac{2\alpha}{m_2}}, \]  

(7)

\[ \omega_{3c} = \omega_{+} (k = 0) = \sqrt{ \frac{2\alpha (m_1 + m_2)}{m_1 m_2}}. \]  

(8)

The wave number expressions within these frequency limits are given by inverting relation (5).

For \( 0 \leq \omega < \omega_{1c} \), the acoustic propagation branch,

\[ k(\omega) = \arccos \left( 1 - \omega^2 \frac{m_1 + m_2}{\alpha} + \omega^4 \frac{m_1 m_2}{2a^2} \right) / a. \]  

(9a)

For \( \omega_{1c} \leq \omega < \omega_{2c} \), the low-frequency band gap,

\[ k(\omega) = \frac{\pi}{a} + i \arccosh \left( -1 + \omega^2 \frac{m_1 + m_2}{\alpha} - \omega^4 \frac{m_1 m_2}{2a^2} \right) / a. \]  

(9b)

For \( \omega_{2c} \leq \omega \leq \omega_{3c} \), the optical propagation branch,

\[ k(\omega) = \arccos \left( 1 - \omega^2 \frac{m_1 + m_2}{\alpha} + \omega^4 \frac{m_1 m_2}{2a^2} \right) / a. \]  

(9c)

For \( \omega_{3c} < \omega \), the high-frequency band gap,

\[ k(\omega) = i \arccos \left( 1 - \omega^2 \frac{m_1 + m_2}{\alpha} + \omega^4 \frac{m_1 m_2}{2a^2} \right) / a. \]  

(9d)

An experimental test of the dispersion relation for the acoustical propagation band is presented in Fig. 2. The frequency-dependent wave phase velocity \( c = \omega/k \) in the acoustical propagation band is obtained by measuring the difference in frequency between two successive resonances of the chain. Indeed, because the chain is of finite length \( L \), it exhibits resonances. Because the last cube of the chain is just connected to the sphere above, the boundary condition at the end of the diatomic chain is considered free. The condition at the excitation boundary is close to a rigid one, as the emitter imposes the acoustic displacement. Consequently, the resonances of this rigid-free resonator are observed at frequencies \( f_m = (2m + 1)c/4L \), with \( m = 0, 1, 2, 3, \ldots \). Measuring \( \Delta f_m = f_m - f_{m-1} \) thus provides a frequency-dependent wave

![FIG. 2. (Color online) Experimental and theoretical dispersion curves for the acoustical propagation branch.](041305-3)
where solutions nonlinear contribution at frequency 2 \( \omega \) source term. The corresponding system of equations of motion 

\[
\begin{align*}
\ddot{u}_n^{nl}(2\omega) &= C e^{-ik(2\omega)an} + D e^{-2k(\omega)an}, \quad (12a) \\
\dot{v}_n^{nl}(2\omega) &= E e^{-ik(2\omega)an} + F e^{-2k(\omega)an}, \quad (12b)
\end{align*}
\]

with constants \( C, D, E \), and \( F \) that have to be found. The previous linear solution [Eqs. (10)] gives the relation between constants \( E \) and \( C \):

\[
E = 2C \frac{1 - \frac{(2\omega)^2}{\omega_c^2}}{1 + e^{ik(2\omega)n}}.
\]

The boundary condition of the semi-infinite granular chain is the absence of second harmonic displacement at the first element of the chain (the excitation boundary); thus \( u_n^{nl}(n = 0) = 0 \). This condition implies that \( D = -C \). A particular solution of the system of nonlinear equations of motion [Eqs. (11)] can then be found in the form

\[
\begin{align*}
\ddot{u}_n^{part}(2\omega) &= De^{2i\omega t - 2ik(\omega)an}, \quad (14a) \\
\dot{v}_n^{part}(2\omega) &= Fe^{2i\omega t - 2ik(\omega)an}.
\end{align*}
\]

The relation between constants \( D \) and \( F \) is then found by substituting this form of solution in the equations of motion (11):

\[
\begin{align*}
\begin{bmatrix} D \\ F \end{bmatrix} &= \frac{1}{\det X} \begin{bmatrix} \Phi \\ \Psi \end{bmatrix},
\end{align*}
\]

with

\[
\begin{align*}
\Phi &= \begin{bmatrix} i \frac{\beta}{2\omega} B & B e^{ik(\omega)an} \sin\left(\frac{2\omega}{2}\right) - \frac{1 + e^{2i\omega t}}{2} \\
\frac{\beta}{2\omega} A & -2B e^{ik(\omega)an} \sin\left(\frac{2\omega}{2}\right) - \frac{1 + e^{2i\omega t}}{2} \end{bmatrix},
\end{align*}
\]

and

\[
\det X = \frac{1 - \frac{(2\omega)^2}{\omega_c^2}}{1 + e^{2i\omega t}}.
\]

In conclusion, the solution for the second harmonic wave in the diatomic chain, valid for the whole frequency range including passbands and band gaps, reads

\[
\begin{align*}
u_n^{nl}(2\omega) &= -De^{-ik(2\omega)an} + De^{-2k(\omega)an}, \quad (16a) \\
u_n^{nl}(2\omega) &= -2D \frac{1 - \frac{(2\omega)^2}{\omega_c^2}}{1 + e^{ik(2\omega)n}} e^{-ik(2\omega)an} + Fe^{-2k(\omega)an}.
\end{align*}
\]

**D. Behavior of linear and nonlinear solutions**

Solutions (10) and (16) are computed for the fundamental frequency and the second harmonic for different fundamental wave frequencies relative to the first cutoff frequency: \( \omega_c/\omega_{1c} \approx 0.36, 0.49, 0.61, 0.79 \), and 1.13. These five fundamental wave frequencies represent distinct regimes of second harmonic generation. A small imaginary part in the form \( k'' = -i\gamma/\omega \) with \( \gamma = 1.5 \times 10^{-4} \ \text{s}^{-1} \) has been added to the wave number \( k \) in order to account for wave dissipation. This produces an overall exponential decay of the linear solutions in fair agreement with the experimental observations presented in the following.

In Fig. 3(a) there exists a first regime \( (\omega_c/\omega_{1c} \approx 0.36) \) where both the fundamental and the second harmonic waves are propagative and where the nonlinear process is weakly influenced by velocity dispersion (a weak influence of the velocity difference between the fundamental wave and the second harmonic). While the fundamental wave exponentially decreases with \( n \), the second harmonic wave amplitude first increases with distance due to the cumulative nonlinear process and then decreases due to dissipation and to the decrease of the nonlinear force (fundamental wave amplitude). This corresponds to the classical case of second harmonic wave propagation.
generation in a dissipative but nondispersive one-dimensional medium [32].

In Fig. 3(b), the excitation frequency $\omega/\omega_{\text{fc}} \simeq 0.49$ is such that the second harmonic frequency is close to (but remains less than) the first cutoff frequency. In this case, the velocity difference between the fundamental and the second harmonic waves can be large: the nonlinear sources are successively in phase or out of phase with the generated second harmonic along the chain due to this larger velocity difference. This manifests itself as oscillations in the second harmonic magnitude as a function of $n$.

In the third case with $\omega/\omega_{\text{fc}} \simeq 0.61$ in Fig. 3(c), the fundamental wave is propagative but the second harmonic is evanescent. Its magnitude quickly increases in the first few grains and then decreases approximately twice as fast as the fundamental wave amplitude. In this case, the second harmonic wave does not accumulate with distance nor does it propagate, it is just forced by the nonlinear sources.

The fourth case with $\omega/\omega_{\text{fc}} \simeq 0.79$ in Fig. 3(d) corresponds to a propagative fundamental wave and a second harmonic wave, but with different propagation modes, acoustical and optical, respectively. The optical propagation mode is due to the diatomic nature of the granular crystal and corresponds to an out-of-phase motion of the adjacent grains. In Fig. 3(d) only the magnitude of the wave is plotted, which does not allow us to observe directly the out-of-phase character between $u_n$ and $u_n^\text{nl}$. This out-of-phase character is however indirectly observed on the second harmonic wave magnitude as a consequence of the nonlinear generation process with an acoustic mode (the fundamental wave) generating an optical mode (the second harmonic wave).

In Fig. 3(e), the last case $\omega/\omega_{\text{fc}} \simeq 1.13$ is plotted. Both waves are evanescent, leading to the attenuation of the waves within the first few grains of the diatomic chain. Still, the second harmonic wave is generated from a nonpropagative fundamental wave.

The experimental results presented in the following section show most of the features described above, within the frequency range where it has been possible to measure the particle motion.

### IV. EXPERIMENTAL RESULTS AND DISCUSSION

We first analyze the frequency content of the received signals after a swept sine excitation of the diatomic granular chain. Transfer functions of the diatomic granular chain, at the excitation frequency and at twice the excitation frequency, are presented; these are compared to the theory and discussed. Then, the amplitude-dependent processes at the excitation frequency (nonlinear resonances and self-action) are analyzed. We finally focus on the exceptionally strong amplitude-dependent effects in the vicinity of branch edges.

#### A. Second-harmonic generation in a diatomic granular chain

In Fig. 4, the time-frequency analysis (spectrogram) of the received acceleration signal and of the emitted signal are presented. Limits of the characteristic frequency bands are schematically shown. The emitted signal, an exponential swept sine of 5-s duration, from 1 kHz to 45 kHz has a quasiconstant amplitude. In contrast the signal transmitted through the diatomic granular chain exhibits variations in amplitude as a function of frequency. If we focus first on the amplitude corresponding to the excitation frequency, it is clear that there exist frequency regions where the fundamental amplitude is very low. These correspond to the band gaps located at 18–28 kHz and above 34 kHz. Also, it is important to notice that when the fundamental amplitude is significant, it is not constant when the frequency is varied. The time-frequency analysis shows some color fluctuations along the fundamental frequency curve, associated with oscillations of the amplitude as a function of frequency. This feature is associated with the presence of resonances in the chain and will be analyzed in the following.

Another important point is the presence of two other curves in the bottom panel of Fig. 4 corresponding to the second and third harmonics of the excitation signal. The amplitudes of the second and third harmonics are weaker than that of the fundamental. However, for a given frequency in the low-frequency band gap, for instance 22 kHz, the second harmonic wave is detected while the fundamental wave is not. This
FIG. 4. (Color online) Time-frequency representation of the excitation signal (top) and the received accelerometer signal (bottom) at the other end of a diatomic granular chain of 40 particles.

manifestation will be analyzed in the following. Finally, it is worth noting that second and third harmonic waves exhibit also some resonances in Fig. 4.

We now analyze in more detail the transfer functions for the fundamental frequency and for the second harmonic obtained from the nonlinear deconvolution method introduced in Sec. I [27].

In Fig. 5, the experimental transfer functions for the fundamental frequency and for the second harmonic are compared to the theoretical solutions of Eqs. (10a) and (10b) and Eqs. (16). For the theoretical fits, a constant magnitude for the excitation displacement as a function of the frequency is taken. When converted from displacement into acceleration, the theoretical magnitude can be compared to the experimentally detected acceleration signal. The theoretical curves exhibit a level increase as a function of frequency at the lowest frequencies. This increase is associated with the $\omega^2$ factor between the excitation displacement amplitude taken independent of frequency, and the received acceleration. For higher frequencies, above 10 kHz typically, there is an overall level decrease, additionally to the band gap structure, originating from the wave dissipation in the system. Actually, in comparison to the wave numbers defined in Eqs. (9) we have added a small imaginary part of the form $-i\gamma\omega$ to account for the viscous-like wave dissipation in the system (here $\gamma = 0.00025$). The experimentally measured static precompression force $F_0 = 4 \text{ N}$ is used for the theoretical evaluation. The experimental noise level here is $-50 \text{ dB}$. As the theoretical solutions are derived for a semi-infinite chain, they do not exhibit resonances like the experimental curves. Except for this difference, good agreement is observed between experiments and theory; the cutoff frequencies coincide well, and the fundamental and second harmonic growth with frequency is well reproduced.

Even the sharp dip of the second harmonic level at $\sim 18 \text{ kHz}$ observed experimentally is reproduced by the model. This effect is related to the case shown in Fig. 3(b), where the velocity difference between the fundamental wave and the second harmonic wave leads to beatings in the second harmonic amplitude either as a function of frequency for a fixed distance or as a function of distance for a fixed frequency.

In the first frequency band gap, it is observed both experimentally and theoretically that the second harmonic amplitude is larger than the fundamental one. This case corresponds to the one of Fig. 3(c) where the fundamental wave is propagative and the second harmonic wave is evanescent but forced by the nonlinear sources that propagate. As a consequence, a second harmonic wave can be detected in this band gap. This is not the case of the fundamental wave when it is generated at a frequency in the band gap. The received level of the latter wave is then below the noise of the experiment. This observation can be interpreted as a nonlinear supratransmission effect. The second harmonic can be transmitted inside the band gap only due to the nonlinear process.

One should notice however two main discrepancies between experiments and theory. First, there is a detected signal in experiments, at frequencies 37–40 kHz both for the fundamental and the second harmonic waves. We do not have a clear explanation for the presence of this signal. It could originate from the presence of a small degree of disorder in the granular chain (which would alter the band structure associated with the perfect crystal), from a resonance effect or noise in the measurement system, or from the excitation of possible rotational modes of propagation [35]. Second, in order to fit correctly the relative levels between the fundamental and the second harmonic contributions, it has been necessary to take a theoretical value for the parameter $\beta$ of quadratic nonlinearity larger than the one predicted by the expansion of the Hertz contact stress-strain relationship (or, equivalently, the absolute value of the excitation displacement amplitude is taken larger than the estimated experimental one). This discrepancy could
be due to the deviation of the real contact (contacting surfaces with microscale asperities) from the macroscopic Hertzian behavior. A similar problem with three-dimensional granular packings is mentioned in Ref. [36], where it is argued that the introduction of a fraction of weaker loaded contacts can explain the relative levels of the second and third harmonics. In our experiment, the microscopic contacts due to surface asperities can play the role of the weaker contacts of three-dimensional packings.

B. Nonlinear resonance frequency shift

In the previous section, the second harmonic generation has been analyzed. The amplitude-dependent effects at the excitation frequency (nonlinear self-action effects) are now analyzed. One way to gain insight into the involved nonlinear processes in this case is to study the resonances of the finite chain, observed in Fig. 5. Focusing on a single resonance of the diatomic granular chain in the acoustical propagation band (Fig. 6), one observes a downward resonance frequency shift and a diminution of the quality factor for increasing excitation amplitudes. This observation is frequent in sandstones, cracked materials, or disordered granular materials [37].

The downward frequency shift can be interpreted as an amplitude-dependent modification of the acoustic wave velocity (averaged over the wave period) or an average softening of the medium (i.e., a diminution of the elastic modulus) [10,39,40]. The longitudinal elastic modulus relative modification, averaged over a wave period, is written \( \langle \Delta E/E \rangle = (E - E_0)/E_0 \), where \( E \) is the longitudinal elastic modulus at a given amplitude and \( E_0 \) is the (linear) longitudinal elastic modulus at vanishing acoustic amplitude [39,40]. The symbol \( \langle \cdot \rangle \) denotes the average over one period of the signal.

The relation between the relative shift in elastic modulus, \( \langle \Delta E/E \rangle \), and the relative shift in resonance frequency, \( \Delta f/f_0 = (f - f_0)/f_0 \), where \( f \) is the resonance frequency at a given amplitude and \( f_0 \) is the (linear) resonance frequency at vanishing acoustic amplitude, can be approximated by \( \langle \Delta E/E \rangle \approx \Delta f/f_0 \) when \( \Delta f/f_0 \ll 1 \). This is the case in Fig. 6, where, at the maximum excitation amplitude, \( \Delta f/f_0 \approx 0.03 \).

The amplitude-dependent elastic modulus \( E = \partial \sigma / \partial \varepsilon \) is obtained from the stress-strain relationship \( \sigma = f(\varepsilon) \) of a given system, where \( \sigma \) and \( \varepsilon \) are the dynamic (acoustic) stress and dynamic strain, respectively.

For a system of two spherical grains in contact, or one sphere in contact with a cube, the Hertzian stress-strain relationship reads [30],

\[
\sigma = -C(\varepsilon - \varepsilon_0)^{3/2}, \tag{17}
\]

where \( \sigma \) is the total tensile stress, \( \varepsilon \) is the acoustic strain, \( \varepsilon_0 \) is the static strain of the contact, and \( C \) is a constant which depends on Young’s modulus and the Poisson ratio of the material as well as grain dimensions and shapes (sphere-sphere or sphere-cube contact). Here a positive strain corresponds to elongation of the system. In the limiting case of a weak acoustic strain compared to the static one \( (|\varepsilon| \ll |\varepsilon_0|) \) relation (17) can be expanded in a power series with the small parameter \( \varepsilon \):

\[
\sigma \approx -C\left((\varepsilon_0)^{3/2} - \frac{3}{2}(\varepsilon_0)^{1/2}\varepsilon + \frac{1}{16}(\varepsilon_0)^{-3/2}\varepsilon^3 + \cdots\right). \tag{18}
\]

The dynamic part of the stress is thus written as

\[
\sigma \approx \frac{3}{2}C(\varepsilon_0)^{1/2}\varepsilon \left[1 - \frac{1}{4\varepsilon_0} \varepsilon + \frac{1}{24\varepsilon_0^2} \varepsilon^2 + \cdots\right]. \tag{19}
\]

Consequently, in the case of a Hertzian interaction law, \( E_0 = \frac{3}{2}C(\varepsilon_0)^{1/2} \) and \( \Delta E/E_0 = (\partial \sigma/\partial \varepsilon - E_0)/E_0 \). This leads to the following relative shift in elastic modulus:

\[
\frac{\Delta E}{E_0} \approx -\frac{1}{4\varepsilon_0} \varepsilon + \frac{1}{24\varepsilon_0^2} \varepsilon^2 + \cdots \tag{20}
\]

Assuming a sinusoidal wave excitation of the form \( \varepsilon = \varepsilon_A \sin \omega t \) and averaging the relative shift in elastic modulus over a wave period gives, at the leading order in \( \varepsilon_A \),

\[
\left\langle \frac{\Delta E}{E_0} \right\rangle \approx \frac{2}{f_0} \Delta f \approx \frac{1}{48\varepsilon_0^2} \varepsilon_A^2. \tag{21}
\]

Equation (22) shows that a Hertzian contact in the weakly nonlinear regime \( (|\varepsilon| \ll \varepsilon_0) \) provides a relative frequency shift \( \Delta f/f_0 \propto \varepsilon_A^2 \) at the leading order in \( \varepsilon_A^2 \). This is expected because the Hertzian nonlinearity in the weakly nonlinear regime leads to a classical power law nonlinearity, known to contribute to resonance frequency shift at the cubic order of the stress-strain relationship. A cubic-order nonlinearity provides \( \Delta f/f_0 \propto \varepsilon_A^3 \). Such behavior has also been predicted recently using the harmonic balance method in a Hertzian granular chain [22]. In Fig. 7(a), the relative frequency shift of the resonance plotted in Fig. 6 is shown; the relative shift in inverse quality factor \( \Delta (1/Q) = 1/Q - 1/Q_0 \) where \( Q \) is the quality factor at a given excitation amplitude and \( Q_0 \) is the (linear) quality factor at vanishing excitation amplitude is shown in Fig. 7(b).

Note that the amplitude-dependent quality factor cannot be estimated precisely by measuring the width of the resonance curve at half amplitude (or -3 dB) when the resonance curve becomes asymmetric. We make use of the following relation to estimate the amplitude-dependent attenuation (which uses the strain amplitude at the resonance instead of the width of the resonance curve) [38,39]:

\[
\Delta \left( \frac{1}{Q} \right) = \frac{1}{Q_0} \left( \frac{V\varepsilon_0^3}{V_0\varepsilon^2} - 1 \right), \tag{23}
\]
relationship of the contacts with longitudinal acoustic excitation. Mechanical hysteresis exists in several developed and widely used contact models in longitudinal solicitation such as the Johnson-Kendall-Roberts model, the Derjaguin-Muller-Toporov model, or the Maugis model [43]. All these models involve adhesion processes which produce asymmetry between the tensile behavior of the contact and its compressional behavior [30,43]. The adhesion processes can arise from a combination of different contributions such as the van der Waals force, the magnetic force, the electrostatic force, capillary forces [30,43], etc. In turn, the paths taken by the stress-strain relationship are not the same for increasing or decreasing contact strain and can be strongly nonlinear. These mechanical hysteretic stress-strain relationships can be approximated in acoustics, in the limit of small wave amplitudes, by the hysteretic quadratic nonlinearity [10]. Consequently, we conclude that the observed effects can be associated here with nonlinear hysteresis of the normal contact interactions originating from the adhesion processes. In other words, the hysteretic nonlinearity of the normal contact interaction is dominant over the cubic nonlinearity of the stress-strain relationship (the weakly nonlinear Hertzian nonlinearity) for the effect of nonlinear resonances.

One should note however that a small deviation from the predicted linear behavior (associated with the quadratic hysteretic nonlinearity) of the inverse quality factor shift as a function of resonance amplitude is experimentally observed in Fig. 7(b). This small deviation from a linear shift can be attributed to the additional contribution of classical dissipation together with classical quadratic nonlinearity, or other combined nonlinear processes leading to nonlinear attenuation [44].

We show in the following that the relatively weak nonlinear effects on resonances reported in this section (a 3% relative frequency shift) can be at the origin of exceptionally strong amplitude-dependent effects at the frequency band edges of the granular crystal.

**C. Nonlinear self-action effects at the band edges**

At the band edges of a phononic crystal, strong frequency-dependent effects occur. The transition from a propagative band to a band gap takes place in a narrow frequency range and is associated with a variation of the acoustic transfer function amplitude by several orders of magnitude, such as in Fig. 5. Even a relatively small softening effect as observed in the previous section on the resonances of the chain can consequently lead to strong amplitude-dependent effects at these band edges. Note that strong amplitude-dependent effects can be observed in similar situations where weak nonlinear effects take place for interfering waves: close to an antiresonance like in Ref. [45] or when two modes of propagation in a medium supporting Biot waves interfere [46], for instance.

The softening is associated with a decrease of the effective modulus of the chain when increasing the wave amplitude. It leads to a decrease of the average velocity and hence a decrease of the wavelength. As the phononic effects including the location of the band edges depend on the ratio between the wavelength and the spatial periodicity of the crystal, a decrease of the wavelength at a given excitation frequency has the effect...
of shifting the band structure toward lower frequencies; i.e., the cutoff frequencies are down-shifted with increasing excitation amplitude. In Fig. 8, the three experimental cutoff frequencies of the diatomic granular chain are plotted as a function of the excitation level. These experimental characteristic frequencies are defined as the frequencies for the mid-level between the band gap level (noise of the experimental setup) and the level of the closest passband. It is observed that all these three characteristic frequencies diminish with increasing excitation level, which is consistent with the nonlinear softening effect.

For the diatomic crystal band structure, there are two passbands, two band gaps, and three cutoff frequencies. A downward shift of the cutoff frequencies can lead to essentially different effects depending on which side of the frequency bands is considered.

In Fig. 9, experimental levels of the acoustic transfer function averaged over 500 Hz at the three band edges, for increasing excitation levels. The detected level is normalized by the excitation level. A linear behavior of the granular crystal would exhibit horizontal lines. For the right edges of the propagation bands (both acoustical and optical), the normalized level decreases with the excitation level. For the acoustical band cutoff frequency, a $-13 \, \text{dB}$ level decrease is observed for a $+16 \, \text{dB}$ increase in the excitation level. In other words, while the excitation level is increased by 16 dB, the received level is only increased by 3 dB. In contrast, for the low frequency cutoff of the optical propagation band, the 16 dB increase of the excitation level provides a 21 dB increase of the detected wave level.

In summary, two essentially different and strong nonlinear self-action effects can be observed at the band edges due to their downward shift with excitation amplitude. We observe a self-attenuation for the acoustical band edge and the optical band right edge (at the optical band high-frequency cutoff) and a self-amplification (also denoted as a self-induced transparency effect) for the optical band left edge. These effects are well explained by the nonlinear softening effect associated here with the hysteretic nonlinearity of normal contacts.

V. SUMMARY

The process of second harmonic generation in a diatomic granular chain has been experimentally and theoretically studied. The rich observed effects associated with the peculiar dispersive properties of such diatomic granular crystals and the nonlinearities are in good agreement with the developed model for the second harmonic generation. Our results provide experimental evidence for the transmission of an evanescent second harmonic wave (with a frequency inside the band gap of the granular crystal) when it is forced by the propagative nonlinear sources (fundamental wave). Effects of the velocity dispersion on the second harmonic generation process are also observed and interpreted.

Besides, we observed and analyzed another nonlinear effect occurring at the excitation frequency. This effect has a nature similar to that of the nonlinear resonance frequency shift observed in mesoscopic materials (rocks, damaged materials, etc.). It is associated with the softening of the medium.
when the excitation amplitude is increased. For a phononic crystal, exhibiting a transmission coefficient with a strong frequency dependence, in particular at the band edges, a small amplitude-dependent frequency shift (equivalent to a softening of the medium) leads to dramatic self-attenuation or self-transparency effects at a given frequency. In other words, the characteristic frequencies of the phononic filter change as a function of the wave amplitude. Hysteric nonlinearity of normal contact interactions is found to be the cause for these observed effects.

These observations and interpretations will be valuable for developing an understanding of nonlinear properties of phononic crystals. It is expected that the effects reported here will lead to applications in wave tailoring by phononic crystals. The unusually strong amplitude-dependent effects of wave attenuation or transparency with increasing excitation amplitude could be used for acoustical devices analogous to the widely found electrical devices. These results provide also a first step toward understanding nonlinear effects in higher-dimension (granular) phononic crystals.

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