

## Research paper

## Enhancement of harmonics generation in hysteretic elastic media induced by conditioning

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## ARTICLE INFO

## Article history:

Received 24 February 2016

Revised 6 August 2016

Accepted 14 September 2016

Available online 15 September 2016

## Keywords:

Hysteresis and conditioning

Nonlinear acoustics

Harmonics

Multistate models

## ABSTRACT

The physical origin of harmonics generation in non classical (hysteretic) elastic media and the mechanisms of energy transfer among harmonics are still not completely understood. Furthermore the well known conditioning effect observed in such materials is known to have a significant influence on the elastic response of consolidated granular media and damaged composites and metals. Here, we show that the elastic non linearity of samples belonging to these two categories increases after having been excited with a relatively low amplitude stress. The observed behaviours could be described by activation features intrinsically present in phenomenological multistate models proposed in the literature.

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## 1. Introduction

Non classical elastic solids [1,2], such as rocks [3], concrete [4], damaged bones [5], metals or composites [6,7], etc., when excited with low amplitude waves (strains between  $10^{-7}$  and  $10^{-5}$ ) manifest typical non linear effects, such as higher order harmonics generation [8,9], break of the proportionality between input and output [10] and of reciprocity [11], amplitude dependence of the damping coefficient [12], generation of sidebands [13] and/or subharmonics [14]. Also, they exhibit memory effects, such as conditioning and relaxation [15,16], i.e. a change in the elastic state induced by the propagating wave which is mostly characterizing their non classical / hysteretic behavior. Conditioning and relaxation are often monitored experimentally by measuring the resonance frequency of a given sample [17] which drops down (softening), after the sample has been excited by a large amplitude stress, and slowly in time recovers back to its original value [18,19].

In all these cases the physical mechanisms responsible for conditioning and for the transfer of energy from fundamental to odd and even higher order harmonics are not clear. Selective non linear absorption phenomena or transparency [20] and activation of non linear features induced by conditioning [21] or thermally [22] might take place, as a consequence of the presence of different physical features at the microscopic scale. These features could be dislocations in metals [23], grain contacts in sandstones and concrete [24,25], cavitation in water layers present at interfaces [26], etc. Often different processes might act at the same time and perhaps affect the behavior in different strain ranges [27,28].

The existence of a link between conditioning and non classical elastic response has been discussed in previous papers [15,29], where the variation of the linear elastic modulus as a consequence of a transition to a non equilibrium elastic

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state has been carefully demonstrated. However, not much attention has been yet devoted to study the changes induced by an applied strain not larger than  $10^{-6}$  in generation of harmonics, or more generally the effects of conditioning on the nonlinear elastic coefficients [30].

Understanding the phenomenology of conditioning in full details is of course of great importance from a physical point of view. But also it could be relevant for applications. In fact, nonlinear ultrasound have been widely used as a tool for damage and defects diagnosis, identification and localization [31–34]. However, in media presenting hysteresis the nonlinear response due to a crack, delamination or imperfect grain boundary could be highly different if the measurement is performed on a relaxed or on a conditioned sample, thus some a priori knowledge of the effects due to memory is necessary for a reliable interpretation of the data.

In this paper, we have considered a granular consolidated material (concrete) and a damaged composite metal, representing two classes of materials exhibiting hysteretic elastic behavior. Their elastic responses to a sweep excitation have been analyzed to investigate whether the low amplitude strain due to the sweep affects harmonics generation, besides inducing the known softening effect (reduction of the linear elastic modulus). The experimental set-up and procedures will be described in the next Section. Results, indicating an anomalous behavior in harmonics generation are discussed in Section 3. Finally, we will show how nonlinearity activation could be intrinsic in existing multistate models [35–39]. In particular, in Section 4 it is shown how the Granato–Lucke model of dislocations [35] could qualitatively predict the observed phenomenology.

## 2. Experimental details

### 2.1. Samples

As mentioned in the Introduction, both samples with damage (e.g. composites or metals) and samples with an intrinsic granular structure (rocks, bones or concrete) exhibit anomalous (hysteretic) elastic behaviors. Here, to highlight the generality of the results presented, we have considered two materials belonging to the two categories, characterized by very different constituents and chemical composition.

The first sample considered was a thin composite metallic plate ( $135 \times 20 \times 3 \text{ mm}^3$ ) with the following composition: Titanium (90%), Aluminum (6%), Vanadium (4%), Iron ( $\leq 0.25\%$ ) and Oxygen ( $\leq 0.2\%$ ). The density of the material was measured to be  $4410 \text{ kg/m}^3$  with a Young modulus 126 GPa and Poisson ratio 0.305. Due to the hardness of the sample, one can expect the integrity of the plate to be affected by the rolling and cutting procedures. The fatigue induced damage was not detectable at visual inspection and the mechanical properties of the sample were not affected by the damage process.

The second sample was a cylindrical concrete sample, with radius 4cm and length 16cm. Its composition was: cement CEM II A-L 42.5R ( $\approx 17\%$ ), sand ( $\approx 38\%$  with grain sizes from 0 to 3 mm radius), gravel ( $\approx 36\%$ , with big grains up to 1.5 cm size) and water to cement ratio 0.5 ( $\approx 8\%$  water). For this sample, aging effects are to be considered since it has been manufactured 5 years before the experiments and it has been kept undamaged.

### 2.2. Experimental set-up.

The generation/acquisition system was composed of an arbitrary waveform generator, connected to a linear amplifier (200x) and an acquisition card (3.25 MHz sampling frequency and 16bits dynamic resolution). The amplifier and cards were connected to the ultrasonic source and receiver sensors, glued to the sample with a thin layer of Phenyl–Salicylate.

In the case of the metal composite sample, two piezoelectric disks (Ferroperm transducers PZ27 with diameter 20 mm and thickness 2 mm, having a flat response in the kHz range) were glued on the upper surface of the plate, acting as emitter and receiver. A sweep source signal, of duration 15 s and covering the frequency interval between 5800 and 6400 Hz, was generated. In that frequency range, the transducers work in radial mode and, given the sample geometry, excite the third flexural mode of the plate, numerically estimated to be at 6180 Hz.

The geometry of the concrete sample, forced us to excite longitudinal waves. Two piezoelectric transducers (with diameter of approximately 10 mm, having a flat response in the kHz range) were glued on the two bases of the cylindrical sample, acting as emitter and receiver. A sweep source signal, of duration 1s and frequency interval between 8800 and 9300 Hz, was generated. In that frequency range, the transducers work in compressional mode. Knowing the sample geometry and mechanical properties, one can easily find that the excitation frequency domain covers the first longitudinal mode.

Linearity of the generation/acquisition system has been carefully tested using a reference linear (undamaged) steel sample with geometrical characteristics similar to that of the composite metallic plate. The coupling (Phenyl–Salicylate) was tested in the frequency range between few kHz to hundreds of kHz and showed a linear behavior up to 0.75 V excitation amplitude with radial transducers and up to 1.25 V excitation amplitude with longitudinal transducers. In the experiments reported in the following, the linearity limit was never exceeded.

### 2.3. Experimental procedure

As mentioned sweep signals were used for the analysis of both samples. The sweep was built with a linear variation of the frequency, resulting in a signal in the form:

$$v(t) = A^{inp} \cos\left(\left(\omega_{min} + \frac{\omega_{max} - \omega_{min}}{T}t\right)t\right) \quad (1)$$

Here  $t$  is time and  $A^{inp}$  represents the signal amplitude. The frequency thus increases from  $\omega_{min}$  to  $\omega_{max}$ . The linear resonance frequency of the investigated mode was within the considered frequency interval.  $T$  is the sweep duration, assumed to be sufficiently long to ensure quasi-standing wave conditions ( $T = 15$  s and  $T = 1$  s for the composite metal plate and the concrete cylinder, respectively).

The experiment was performed repeating the sweep injection varying the excitation amplitude. For each amplitude a signal  $v_i(t)$  was recorded by the acquisition card and analyzed as it will be discussed in the next Section. Two slightly different acquisition procedures were adopted for the two samples:

- In the case of the composite metal plate, 13 increasing amplitudes, from 0.04 V to 0.64 V (before amplification) were considered, i.e. up to maximum strain of  $5.5 \cdot 10^{-5}$ , measured in the center of the plate using a laser doppler vibrometer. This first set of measurements will be denoted in the following as measurements on the unconditioned sample. Once the largest amplitude was reached, the very same sweep at 0.64 V was injected repeatedly for 20 min to condition the sample. As a consequence, a drop of 15 Hz of the resonance frequency at the lowest excitation amplitude, taken as a reference, was observed, corresponding to a softening of the material, in agreement with other observations in the literature [15]. Afterwards, the 13 measurements at increasing sweep amplitudes were repeated again, to analyze the nonlinear properties of the conditioned sample. This set will be denoted in the following as measurement on the conditioned sample. It was preliminarily tested that full conditioning was obtained in the 20 min duration of application of the large amplitude excitation. Also, it was verified that the effect of conditioning, i.e. the resonance frequency drop, is fully reversible, i.e. if the sample was unperturbed, on a time scale of the order of hours, the resonance frequency returns back to its original (unconditioned) value. During the relaxation process, we have repeated at different relaxation times the 13 measurements at increasing amplitude to monitor the relaxation of the nonlinear modulus, as it will be discussed in the next Section.
- In the case of the concrete cylindrical sample, measurements were performed at increasing excitation amplitude from 0.01 V to 1.0 V (before amplification), with 11 successive measurements. Afterwards, to analyze conditioning induced by the same sweeps used for probing, additional 10 measurements were immediately performed decreasing amplitude from 1 V to 0.1 mV. The two sets of measurements will be denoted as upgoing and downgoing. Note that each measurement is performed with increasing frequencies during the sweep.

The experiments were repeated more than once on each sample, for the same mode and for other modes as well. Results obtained were repeatable from a qualitative point of view. In the following only one set of data for each sample is discussed.

## 3. Results and discussion

### 3.1. Nonlinearity

#### 3.1.1. Data analysis

As remarked in the previous Section, the experimental campaign allowed us to have several sets of signals for different amplitude variation protocols:

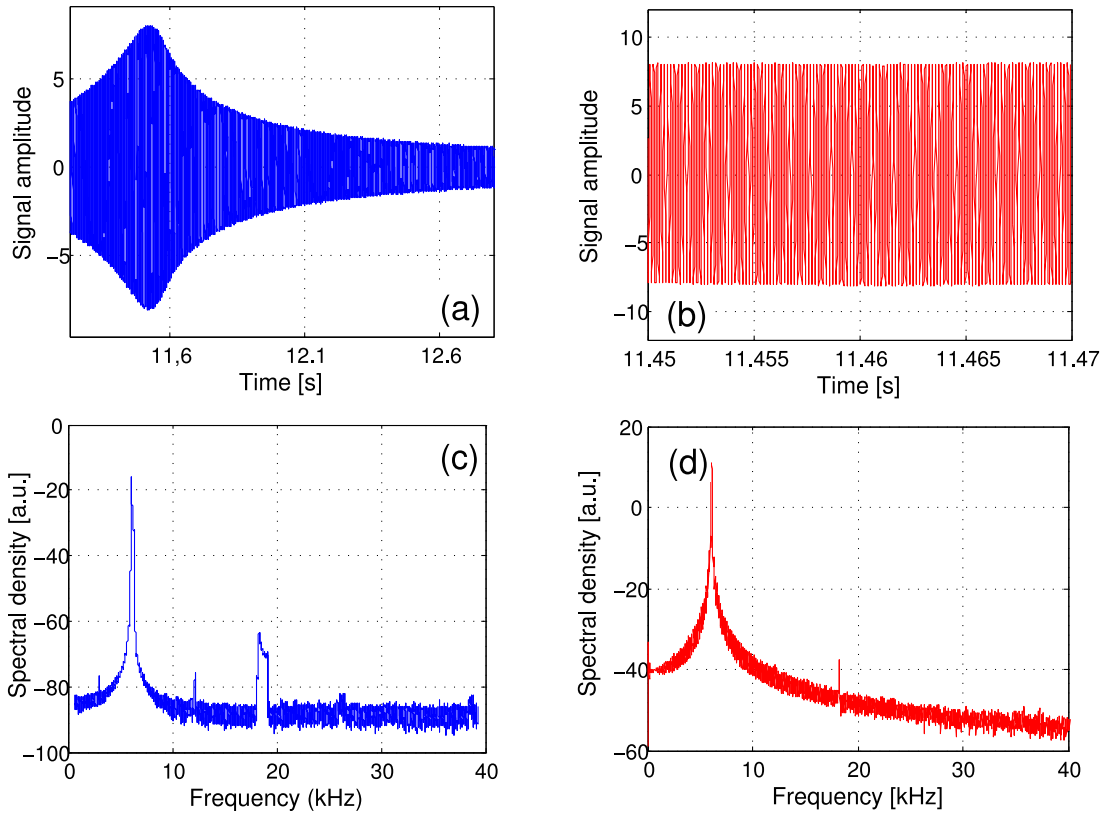
- sets of 13 signals at increasing amplitude for the composite metal plate in the unconditioned and conditioned state and at different relaxation stages, i.e. at different time instances during relaxation (with a total of four sets of data);
- a set of 11 signals at increasing amplitude (i.e. during the conditioning phase for the material) and a set of 10 signals at decreasing amplitude (i.e. for the sample conditioned by the probing wave itself), in the case of the concrete sample.

The frequency spectrum of each signal of each series was analyzed. A typical example of signal and related Fourier Transform (FFT) is shown in Fig. 1a and c. The signal was recorded on the unconditioned composite metal plate at an intermediate amplitude of excitation. For each amplitude  $A_i^{inp}$  of excitation, the highest maximum in the spectrum allows to identify the resonance frequency of the sample  $\omega_{r,i}$  and its corresponding amplitude  $A_i$ .

The relative variation of resonance frequency with respect to the linear value  $\delta f/f = (\omega_r - \omega_0)/\omega_0$  could be calculated. Here  $\omega_0$  is the linear resonance frequency, i.e. the resonance frequency in the limit of excitation amplitude going to zero. In practice,  $\omega_0$  is the sample resonance frequency measured at the lowest excitation amplitude. Also, an estimate of nonlinear attenuation could be given by the quantity:

$$\gamma = A_i/A_i^{inp} - A_0/A_0^{inp} \quad (2)$$

Here the amplitude  $A_0$  is measured for the lowest excitation amplitude  $A_0^{inp}$ , i.e. expected as a linear response.



**Fig. 1.** Experiment on the unconditioned composite metallic plate excited with a sweep at excitation amplitude  $A = 0.35V$ . (a) time window of the signal used for the analysis of Section 3.1.; (b) shorter time window of the signal in (a) close to the maximum amplitude of the signal; (c) spectrum of the signal in (a); (d) spectrum of the signal in (b). The time window used in subplot (b) could be swept along the full signal in order to get the spectrum for each portion of the signal in (a), as discussed in Section 3.2.

Finally, second and third order harmonics emerge from noise and their amplitudes ( $A_{2\omega_i}$  and  $A_{3\omega_i}$ , respectively) could be measured. At low amplitudes of excitation (particularly in the case of the concrete sample), harmonics are within the noise level and thus not detectable. At higher amplitudes third harmonics are more significant in amplitude than second harmonics.

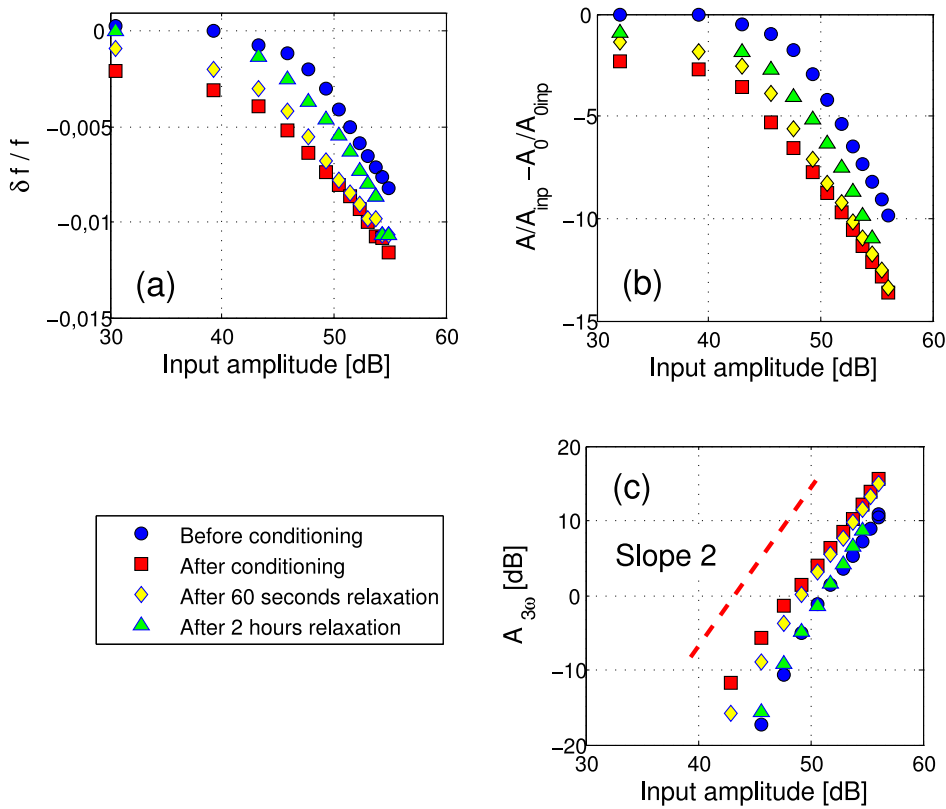
### 3.1.2. Results

In Figs. 2 and 3, the variation of the resonance frequency (subplot(a)), the parameter  $\gamma$  (related to attenuation, Eq. (2), see subplot (b)) and the third harmonic amplitude (subplot (c)) are shown as a function of the excitation amplitude, for composite metal and concrete, respectively.

In the case of the composite metal plate, for both the unconditioned and the conditioned states, the resonance frequency drops with increasing amplitude of excitation (the maximum frequency drop is almost 1%). As already remarked, immediately after conditioning the resonance frequency does not return to its original value (compare the red and blue symbols at the lowest excitation amplitude in Fig. 2a). The total drop due to conditioning is of around 15 Hz. However, repeating the measurement at successive intervals of time the curve tends to recover the same values as before conditioning. The same qualitative behavior is also observed for the attenuation parameter  $\gamma$  (subplot(b)).

The third harmonic amplitude (Fig. 2c) increases up to about 1% of the fundamental. The dependence on amplitude of excitation is a power law  $A_{3\omega} = aA_{inp}^b$ , which in log-log scale appears as a straight line. The power exponent is close to 2, in agreement with literature observations reported for other hysteretic elastic solids. The effect of conditioning is a significant increase of the third harmonic generation, without changing significantly the slope (compare red squares with blue circles), although a slight decrease of the slope could be appreciated. Repetition of the experiment during relaxation indicates the increase to be slowly reversible: after full relaxation of the sample (which lasts for a few hours) we notice approximately the same results as for the not conditioned sample (cyan triangles).

Similar results are found for the concrete sample (Fig. 3). In this case, conditioning is induced by the wave itself, thus it is expected to be less strong than in the previous case, in which a 20 min conditioning at the largest amplitude of excitation was applied. Nevertheless, some differences could be appreciated between the branches of the curves obtained increasing (blue circles) or decreasing (red squares) amplitudes. As in the previous case, in the decreasing branch, the sample is slightly



**Fig. 2.** Measurements performed on the composite metal plate. (a) Resonance frequency shift as a function of excitation amplitude; (b) Attenuation parameter  $\gamma$  (see Eq. (2)) as a function of excitation amplitude; (c) Third harmonic amplitude as a function of excitation amplitude. The red dashed line corresponds to a line (power law behavior) with slope 2. Data refer to measurements performed before and after conditioning. Also the measurement was repeated during relaxation after few seconds and after 2 h during which the largest amplitude stress was not applied.

conditioned by the largest strain induced during the increasing excitation amplitude analysis. As a consequence, for the same level of excitation the resonance frequency is slightly smaller. The effect of conditioning seems to be negligible on the attenuation parameter and generation of harmonics. The latter could however be expected. Indeed, as visible in Fig. 2c, the recovery for harmonics generation is faster than for the resonance frequency. In the case of the concrete sample, partial relaxation between successive measurements is occurring, since at least 1 min occurs between two successive acquisitions.

### 3.2. Harmonics generation

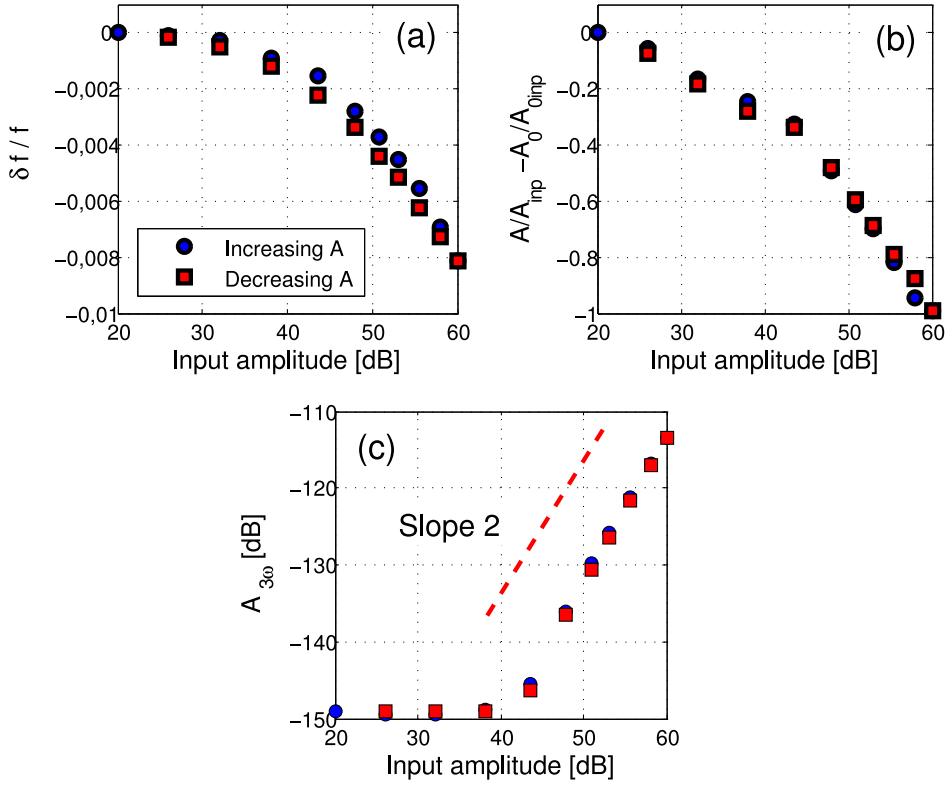
#### 3.2.1. Data analysis

The spectrum of the recorded signals shown in Fig. 1c was obtained analysing the full signal. As expected, the plot shows a peak in correspondance of the third harmonic, i.e. at a frequency  $\omega = 3\omega_r$ , where  $\omega_r$  is the frequency corresponding to the maximum in the spectrum. Such a peak appears highly deformed, with anomalously larger amplitudes at frequency slightly larger than  $3\omega_r$ . However, as shown in Fig. 1b and d, if only a short time window of the signal is analyzed (i.e. a time window in which the source is at a given almost monochromatic frequency), the spectrum around the third harmonic is peaked, as expected.

Data were thus processed with a moving window Fast Fourier Transform (FFT), to extract, for each frequency component of the sweep excitation, the corresponding amplitudes at the fundamental (same frequency as the source) and its harmonics. The procedure is the following:

- for each time  $t$ , the signal  $w_i(t)$  is defined by making a smoothened rectangular window of  $v_i(t)$  in the interval  $[t - \Delta t, t + \Delta t]$ , with, e.g.,  $\Delta t = 12$  ms, in the case of the composite metal plate;
- the frequency  $\omega_i(t)$  and amplitude of the fundamental  $A_{1,i}(t)$  (first peak in the FFT spectrum of  $w$ ) are recorded for each time, together with the amplitudes of the second and third harmonic peaks  $A_{2,i}(t)$  and  $A_{3,i}(t)$ ;
- the detected amplitudes are plotted vs. time, which is equivalent to plot them vs. input frequency, since in the sweep source the input frequency increases linearly with time.

The analysis is performed only in a small time range around the time at which the peak in the response is reached (i.e. in a narrow frequency range around the resonance frequency).



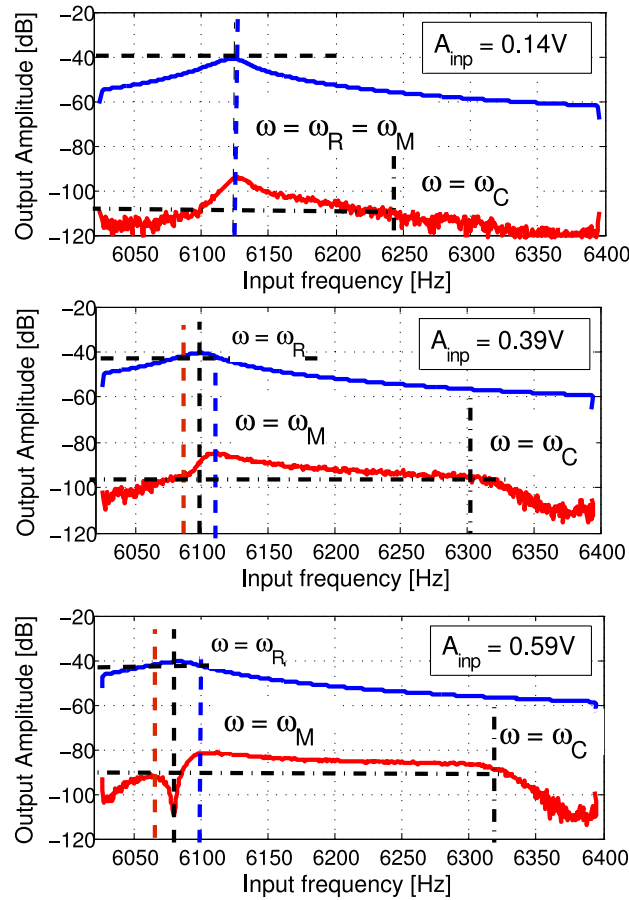
**Fig. 3.** Measurements on the cylindrical concrete sample. (a) Resonance frequency shift as a function of excitation amplitude; (b) Attenuation parameter  $\gamma$  (see Eq. (2)) as a function of excitation amplitude; (c) Third harmonic amplitude as a function of the excitation amplitude. The red dashed line corresponds to a line (power law behavior) with slope 2. Data refer to measurements performed with increasing and decreasing excitation amplitude tests.

### 3.2.2. Results

We first analyze the case of the composite metal plate in the unconditioned state. Results for three selected (low, intermediate and large) excitation amplitudes are reported in Fig. 4, in the three rows respectively. We can observe that:

- (a) softening: the maximum of the fundamental occurs at an input frequency which decreases with increasing amplitude of the excitation (see the position of the maximum of the blue curves). This effect is equivalent to the softening of the sample observed in standard resonance frequency experiments [17] and discussed in Fig. 2a;
- (b) third harmonic generation (small and intermediate amplitudes): the behavior of the third harmonic vs. input frequency (red curves) presents features common to all excitation amplitudes and anomalous effects which are strongly amplitude dependent. In all cases, the third harmonic amplitude increases with increasing the amplitude of the fundamental, up to reaching the peak value. However, after having reached its peak, the energy of the third harmonic does not decay rapidly to zero with time (or frequency), as it would have been expected if the nonlinear modulus was not affected by conditioning. We could also notice that the peak of the third harmonic energy occurs at a frequency  $\omega_M$  which is slightly larger than the resonance frequency of the sample ( $\omega_R$ ). The difference between the two frequencies increases with amplitude: notice the difference between the vertical dashed black and blue lines. As mentioned, after the maximum is reached, the amplitude of the third harmonic decays very slowly, denoting an anomalous increase of harmonics generation. The effect is more and more pronounced increasing the amplitude of the driving. Standard decay due to attenuation is always at frequencies larger than a cut-off frequency  $\omega_C$  of about 6310 Hz (except at the smallest amplitude of the excitation). This region corresponds to the decay to zero of the third harmonic amplitude to the right of the dashed-dotted black vertical line. The third harmonic amplitude at  $\omega = \omega_C$  is  $A_{3,C}$  and corresponds to the amplitude of the third harmonic at a frequency  $\omega_D < \omega_R$ , where a discontinuity in the harmonic generation is observed: the vertical dashed red line indicates  $\omega_D$  where a sudden change in harmonics generation is noticed. Finally, we could observe also that at the frequency  $\omega_D$  the amplitude of the fundamental is roughly the same as that at  $\omega = \omega_M$ ;
- (c) anomalous attenuation at large amplitudes: the behavior at larger amplitudes of excitation is similar except in the frequency range close to resonance (between the red and the blue vertical lines), where a different behavior in the generation of the third harmonic emerges. We observe an anomalous absorption of the third harmonic, which presents a minimum in correspondence of the maximum of the fundamental. Thus, we can assume that extra absorption is





**Fig. 4.** Measurements on the composite metal plate. Features in the generation of harmonics at low, medium and large excitation amplitudes: fundamental (blue) and third harmonic amplitudes (red) vs. input frequency resulting from a moving window FFT analysis. Frequency is reported on the x-axes, corresponding to time due to the choice of a linear sweep function. The vertical and horizontal lines are guide to the eye to easily detect the interesting features and frequencies at which they occur. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

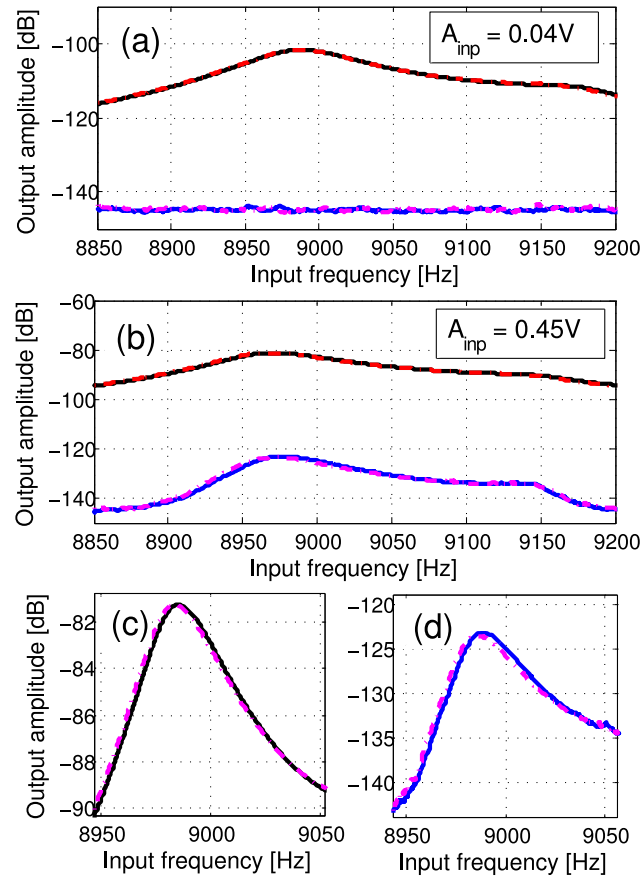
generated, perhaps due to wave mixing of third harmonic and fundamental components, leading to an increase in second harmonic generation, possibly with origin similar to that responsible of self-induced transparency [40].

Similar results to that observed at low and intermediate excitations are observed also in the concrete sample (Fig. 5). In particular the higher generation of third order harmonic after having reached the peak in fundamental is again observable. In the case of concrete, most of the effects are weaker than in the case of the composite metallic plate. The anomalous behavior at larger amplitudes (Fig. 4c) was not observed, perhaps only because strains at play here are smaller than those for the other sample.

Furthermore, in the case of concrete, for each input amplitude two sets of measurements were available: one when the protocol of increasing amplitude was applied and the second when the protocol of decreasing amplitude was applied, i.e. when the sample is (at least partly) conditioned at the largest amplitude of the excitation used in the first protocol. Both results are shown in Fig. 5. Effects of conditioning (i.e. the differences between the two cases) can only be appreciated in the zoom reported in subplots (c) and (d). Albeit with a weak effect, conditioning is still playing a role.

An alternative representation of the results could be adopted. The harmonics amplitudes could be plotted as a function of the fundamental amplitude for any given level of the amplitude of excitation, as shown in Fig. 6 for the composite metal plate. The increase in generation of the third harmonic during the unloading phase of the sweep (i.e. after the peak of the fundamental amplitude is reached), is evident for all three amplitudes considered. Hysteretic loops are formed and the amplitude of the third harmonic emerges from noise at lower values of the fundamental amplitude during unloading than during loading. The same features as discussed in Fig. 4 could be appreciated.

No activation in second harmonic generation seems to be present. For the three amplitudes considered, the second harmonic contribution is always very small (barely out of noise) and the loading/unloading branches are well superimposed.



**Fig. 5.** Measurements on the concrete cylinder sample. Fundamental (black) and third harmonic (blue) amplitudes vs. input frequency resulting from a moving window FFT analysis for selected excitation amplitudes. Frequency is reported on the x-axes, corresponding to time due to the choice of a linear sweep function. Zooms of subplot (b) are reported in subplots (c) and (d) to appreciate differences between measurements obtained during the increasing and decreasing amplitude observations. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Similar results are found in the case of the concrete sample: Fig. 7. Second harmonics are always within noise level, thus have not been reported. The third harmonic is also undetectable for the lowest amplitude of excitation (black line); for the other two amplitudes considered, hysteric loops are formed, similar in shape to those noticed in the case of the composite metal plate at low and intermediate amplitudes of excitation discussed before.

It is interesting to notice the good superposition of the first part of the upgoing branches in the case of concrete. This was not the case for the composite metal sample. This is again an indication of a different conditioning process in the two cases: for concrete, two measurements at successive amplitudes are repeatable in the same range of amplitudes of the fundamental. Thus, the first one is not conditioning the second; in the case of the composite plate, the highest strain reached at a given amplitude is inducing conditioning, so that, for the same values of fundamental amplitude, more harmonics are generated during the second sweep.

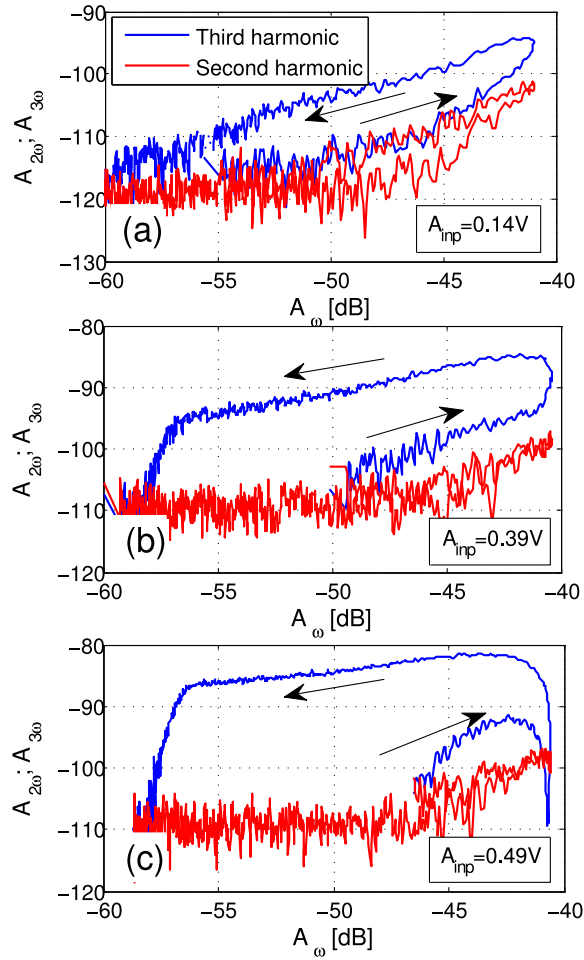
### 3.3. Discussion

Among the results reported, we are mostly interested in the increased rate of harmonics generated after strong conditioning. This is evident from Fig. 2. After having perturbed the composite metal sample with a sequence of strong amplitude sweeps for a sufficiently long time, for the same input level the amplitude of the third harmonic is higher. This effect is similar to that observed in strongly damaged concrete [21].

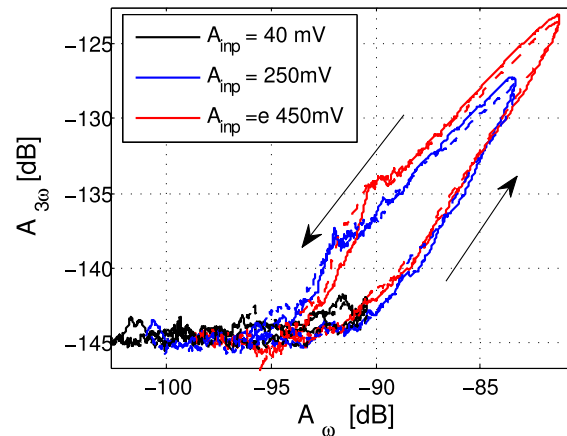
This is not true in the concrete sample, in which decreasing the amplitude of excitation after having reached the maximum strain amplitude the rate of harmonics generation is not varied. However, here the conditioning level (sweep at the largest strain) is active only for a short time. Furthermore, in the case of concrete a significant time interval occurred between successive measurements, hence the mechanisms responsible for eventual activation of generation of harmonics could have been partly/completely relaxed within the duration of the experiment.

Results reported also indicate another conditioning effect. Even when the excitation is still small and of short duration, the propagating wave itself is partially conditioning the sample. As a result, the generation of the third harmonics is more

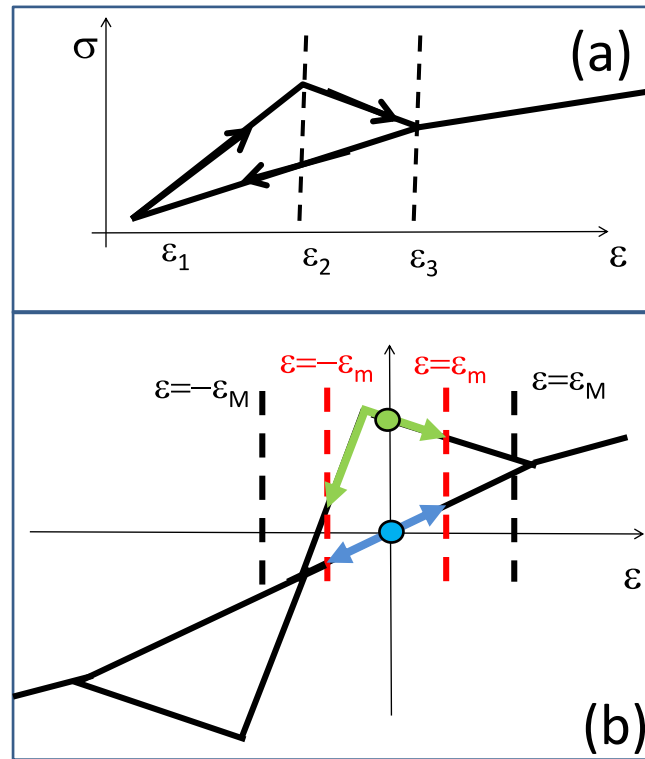




**Fig. 6.** Measurements on the composite metal plate. Second and third harmonic amplitudes vs. fundamental amplitude for different values of the excitation amplitude. Same data as in Fig. 4.



**Fig. 7.** Measurements on the concrete cylinder sample. Third harmonic amplitude vs. fundamental amplitude for different values of the excitation amplitude. Same data as in Fig. 5.



**Fig. 8.** Schematic representation of the dislocations model: a) microscopic equation of state (only the compressive branch is reported); b) conditioning effects on a selected microscopic element. The strain ranges are reported as red (black) vertical lines for the low (conditioning) amplitudes of excitation. The blue (green) circle denotes the initial state of the element before (after) conditioning. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

efficient at frequencies larger than the resonance frequency, i.e. after the peak in the strain distribution within the sample (see Fig. 4 and 5). This activation of non linearity takes place for both samples. Note that the sweep duration is of the same order of magnitude in the two cases, thus it is not surprising to observe similar effects. Such a conditioning effect seems to relax very rapidly. For both samples, third harmonic generation is stronger only shortly after the peak strain (maximum of fundamental) is reached. Within the same sweep, after a short time the rate of harmonics generation (for the same amplitude of the fundamental) returns to be the same before and after conditioning.

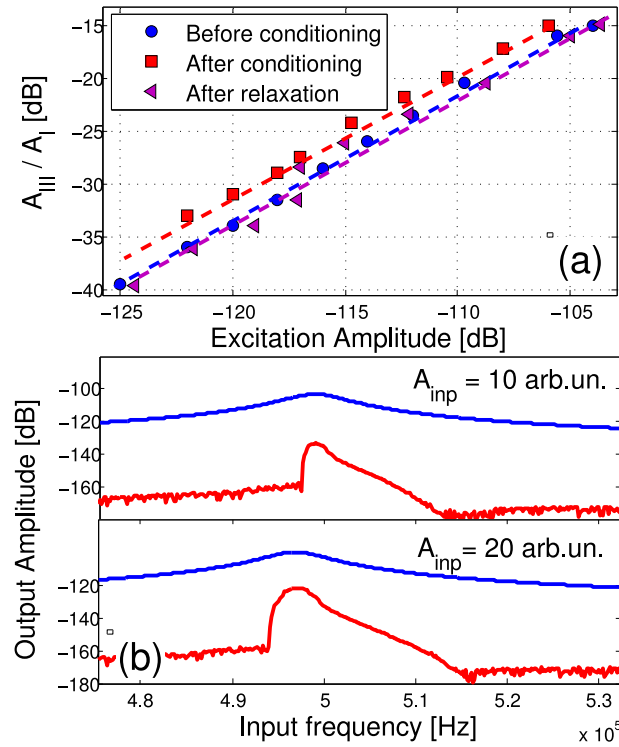
Finally, we wish to note that in several cases generation of subharmonics was also observed [14,32,41] with a very rich phenomenology. In our case, we did not observe evidence of subharmonics in the detected signals. However, it might be that strains at play in our case are much smaller than the threshold of energy required for their generation. It could be of interest to analyze also the effects of conditioning on sub and super harmonics generation.

#### 4. Theory

As already mentioned, the origin of non classical nonlinearity has to be traced back to microscopic features. However, several mechanisms could be thought: internal friction [42], clapping [43], dislocation dynamics [23,27], adhesion [37,44], interstice properties at the grain boundaries [45], etc. Since the analytical description of the multivalued equation of state needed to produce hysteresis and memory effects is difficult, often phenomenological multistate models are introduced [37–39].

Generally, in these models the macroscopic behavior of a nonlinear elastic solid is obtained through a statistical average of simple microscopic stress-strain relations. These relations are defined as a sequence of linear stress-strain equations which are valid in different strain intervals. The goal of this Section is to show that the phenomenon of activation in harmonics generation could be easily introduced or be already implicitly present in such models.

To this purpose, we have applied the Granato–Lucke model [35]. A dislocation line interaction with a point defect is described as follows (see Fig. 8). Increasing the compression, at a certain strain  $\epsilon_1$  the interaction starts taking place, resulting in an increase of the stress with respect to the expected linear value. At a larger strain level ( $\epsilon = \epsilon_2$ ), the dislocation line encompasses the defect, with a consequent decrease of the stress with further increase of strain. Finally, at  $\epsilon = \epsilon_3$  dislocations are no longer interacting with the point defect and thus the stress increases again. During the unloading phase, there is no further interaction with the point defect.



**Fig. 9.** Numerical results of the effects of conditioning using the theoretical model of Fig. 8. (a) Amplitude ratio of the third harmonic over the fundamental before and after conditioning. Results after full relaxation are also shown. Compare with experimental results in Fig. 2c. (b) Third harmonic amplitude vs. frequency obtained using a moving window FFT analysis for low and intermediate amplitudes of excitation. Compare to experimental data in Fig. 4b.

As a result we can define the multi-state constitutive equation as shown in Fig. 8a. Considering an ensemble of nonlinear features, it is reasonable to assume that different dislocations may break away from the point defects with which they interact at different values of the critical strains  $\epsilon_i$  ( $i = 1, 2, 3$ ). Thus we have a statistical ensemble of elements and the global behavior is obtained averaging over a given distribution of critical strains.

Of course, the equation of state is symmetric around  $\epsilon = \epsilon_1$ , with respect to tension and compression, as shown in Fig. 8b. Given  $\epsilon_1$  to assume values according to a statistical distribution, the equation of state presented, contains intrinsically effects of conditioning for at least some choices of the values of the strain at the transition points. Let us consider the behavior of one non linear element, with  $\epsilon_1 < 0 < \epsilon_3$ . At the initial time  $t = 0$ , its state corresponds to the blue circle in Fig. 3b. If the element is excited at low levels of strain ( $-\epsilon_m \leq \epsilon \leq \epsilon_m$ ), the element does not change state and behaves linearly (blue arrows). If we condition at a larger strain such that  $-\epsilon_M \leq \epsilon_1 \leq \epsilon_M \leq \epsilon_3$ , the element switches to the upper branch of the stress-strain curve and, when strain returns to zero ends up in the state denoted by the green circle. Repeating the experiment at the low strain amplitude, it moves on the upper branch only (green arrows), thus contributing with a negative modulus, resulting in a lower average modulus (if a proper choice of the parameters is ensured), and behaving nonlinearly. Thus, conditioning is expected to induce both softening and nonlinearity activation.

Simulations of the experiment discussed previously have been performed (details about the parameters and simulation technique adopted are omitted for brevity). The results are shown for the third harmonic in Fig. 9a and b (equivalent to Figs. 2c and Fig. 4b). The qualitative agreement with experiments is good: both softening (decrease of the frequency corresponding to the peak of the fundamental) and non linearity activation (increase of harmonics generation at frequencies larger than  $\omega_r$ ) are predicted.

The physical mechanisms used here to model the system are not adequate for describing the behavior at larger amplitudes. However, it is known that increasing amplitude of excitation other mechanisms start playing a role, such as the interaction of dislocations with defects in the glide plane [27], which are not included here. Further work is in progress in this direction, which could also enable to predict the shift between the amplitude peaks of the fundamental and third harmonics measured in experiments and not predicted by the current approach.

## 5. Conclusions

Conditioning a hysteretic elastic material with the application of a strain in the range  $10^{-6}$  to  $10^{-5}$  is known to have significant effects on its elastic properties. In general, a softening effect is observed [18] and in the case of damaged solids

an activation of nonlinearity was noticed [21]. Both effects were linked to variations in the linear elastic modulus. Here, we have shown that an increase in the rate of third harmonic generation is also taking place, in addition to the mentioned effects.

The results presented highlight the influence of conditioning on the nonlinear elastic moduli of the material, which seems to be increased on a conditioned sample, at least in the cases considered here. Furthermore, relaxation of the effects on conditioning on harmonics generation seems to be acting on two time scales. On one side, we have observed a long term relaxation (time scale of the order of hours), taking place when the sample is conditioned for a long time at a large excitation level: see Fig. 6. On the other side, even a short duration, small amplitude excitation, such as that induced by a sweep, is inducing a variation in harmonics generation: see Fig. 4 and 5. The effect is however disappearing in a short time.

These novel observations should be further investigated and confirmed in other samples and other frequency ranges, before impacting the development of theoretical models aiming to introduce physical mechanisms responsible of the transfer of energy from fundamental to higher order harmonics. At least for the short time conditioning (i.e. the increase in third harmonic amplitude after the sample was excited at resonance), the time scale considered could be compatible with thermal relaxation only; on the contrary, thermal effects are, in our opinion, not sufficient to predict the other relaxation time scale observed (of the order of hours).

A phenomenological multi-state approach similar to that discussed here could be a useful tool for suggesting additional experiments. The aim should be to allow individuating different features in conditioning effects due to different microstructural features, as recently proposed to classify the nonlinear response of materials with different form of damage [46,47].

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