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Strain-induced kinetics of intergrain defects as the mechanism of slow dynamics in the nonlinear resonant response of humid sandstone bars

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A closed-form description is proposed to explain nonlinear and slow dynamics effects exhibited by sandstone bars in longitudinal resonance experiments. Along with the fast subsystem of longitudinal nonlinear displacements we examine the strain-dependent slow subsystem of broken intergrain and interlamina cohesive bonds. We show that even the simplest but phenomenologically correct modeling of their mutual feedback elucidates the main experimental findings typical for forced longitudinal oscillations of sandstone bars, namely, (i) hysteretic behavior of a resonance curve on both its upward and downward slopes, (ii) linear softening of resonant frequency with an increase of driving level, and (iii) gradual recovery (increase) of resonant frequency at low dynamical strain after the sample was conditioned by high strain. In order to reproduce the highly nonlinear elastic features of sandstone grained structure a realistic nonperturbative form of stress-strain relation was adopted. In our theory slow dynamics associated with the experimentally observed memory of peak strain history are attributed to strain-induced kinetic changes in concentration of ruptured intergrain and interlamina cohesive bonds, causing a net hysteretic effect on the elastic Young's modulus. Finally, we explain how enhancement of hysteretic phenomena originates from an increase in equilibrium concentration of ruptured cohesive bonds that are due to water saturation.

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Apart from their excellent static characteristics as building materials, sandstones have been shown to demonstrate a number of unexpected and even surprising dynamical properties [1–5]. Here we consider the numerous experimental results on nonlinear resonant response exhibited by sandstone rods in forced longitudinal oscillations that appear even at extremely small forcing levels and consequently at small dynamic strain [1–5]. The most intriguing nonlinear feature is slow dynamics, which are defined here as long-term (minutes to hours) changes of elastic properties in response to a disturbance such as dynamic and static strain or temperature.

Specifically, we have to emphasize that in the vicinity of bar resonant frequency the longitudinal alternating drive produces strong essentially nontrivial nonlinear responses: (1) At high drive levels the effective width of resonance curves depends on the direction of frequency sweep; it is narrower for upward sweeps (i.e., from lower to higher frequencies) than at downward sweeps (i.e., from higher to lower frequencies) [1–5]. This effect proves to be a typical manifestation of slow dynamics and can be treated as hysteresis both on low- and high-frequency slopes of a resonance curve. (2) The resonance peak is shifted toward lower frequency almost linearly with an increase of driving amplitude [1,4]. (3) Other evidence of slow dynamics comprises gradual recovery (increase) of resonant frequency to its original value as defined at extremely low drive level after the sample has been conditioned at a high drive level [3,5].

These facts cannot be understood in the framework of standard theories of resonant nonlinear response [6] and imply memory of peak strain history [2]. Some aspects of the problem have been explained by the interpretation of Guyer *et al.* [7] in the framework of a McCall-Guyer quasistatic model [8]. This approach uses the concept of auxiliary hysteretic elements that allows the introduction of an additional nontrivial nonlinear term in the dynamical equation for the field of longitudinal displacements. However, this theoretical treatment lacks completeness in that it initially neglects the dynamics of hysteretic elements and postulates temporal evolution of the amplitude-frequency characteristic (the key point of claimed results) to be developed afterwards. Although Capogrosso-Sansone and Guyer recently suggested dynamical realization of the McCall–Guyer quasistatic model [9], evaluating its adequacy to explain experimental data turns out to be difficult.

In this rapid communication we omit the idea of auxiliary hysteretic elements as the sole approach for treating all peculiar hysteretic phenomena and call attention to an alternative idea used by Davydov and Ermakov for the description of bistability in nonlinear resonant tunneling of electrons through a set of potential barriers [10]. Their approach consists of explicit but physically motivated separation of a given physical system into two nonlinear subsystems, namely, fast and slow subsystems with mutual coupling taken into account.

For sandstone bars we identify the fast subsystem with the field of rapid longitudinal displacements while the slow subsystem represents the concentration of defects in intergrain contact bonds. In doing this we bear in mind that, because of preferable vapor condensation onto surfaces with greater concave curvature [11], the sandstone pore structure [4,11] retains some residual pore water [11], and its impact on the resonant properties of rock is crucial [12,13]. Thus, thermodynamical estimations applied to porous rocks show that in-

tergrain cohesive forces become weaker in the presence of water [14] that agrees with an alternative conception of swelling pressure [13,15]. This treatment is supported by recent experiments [13] that establish an abrupt decrease in the Young's modulus within the first 20% interval of water saturation (i.e., until the degree that void surfaces become completely wet). We could additionally invoke ordinary capillary forces [13] or hydrolysis of silicon-oxygen-silicon bond chains [16] in our consideration. However, either of these mechanisms also leads to softening of the Young's modulus with an increase of saturation. Here the significant issue is apparently not in excessive (presumably unclaimed) details of all plausible mechanisms that might modify the Young's modulus in a qualitatively similar way, but in their reasonable concise formalization by means of a minimal number of slow fields.

According to Kosevich [17] the equilibrium concentration of defects associated with a stress σ is given by the formula,

$$c_\sigma = c_0 \exp(v\sigma/kT), \tag{1}$$

where k and T are the Boltzmann constant and the absolute temperature, respectively, and the parameter $v>0$ stands for a typical volume that accounts for a single defect and characterizes the intensity of dilatation. The equilibrium concentration of defects in an unstrained bar c_0 has to be some function of both temperature T and water saturation s. In order to describe strain-induced changes in nonequilibrium concentration of defects c, we assume that at any instant of time t the concentration c must evolve to its would-be equilibrium value c_σ, where the stress σ in Eq. (1) is applied at the same instant. Supposing the distributions of activation barriers for defect annihilation U and activation barriers for defect creation W are uniform, respectively, over the ranges $U_0 \leqslant U \leqslant U_0+U_+$ and $W_0 \leqslant W \leqslant W_0+W_+$ with U_0, U_+ and W_0, and W_+ being insensitive to the choice of bar cross section, then we will deal with the density of defect concentration g governed by the following kinetic equation:

$$\partial g/\partial t = -[\mu\theta(g-g_\sigma) + \nu\theta(g_\sigma - g)](g-g_\sigma). \tag{2}$$

Here $\mu=\mu_0 \exp(-U/kT)$ and $\nu=\nu_0 \exp(-W/kT)$ are the rates of defect annihilation and defect creation, respectively, $g_\sigma=c_\sigma/U_+W_+$, and $\theta(z)$ designates the Heaviside step function. The quantities g and c are related by the simple definition

$$c = \int_{U_0}^{U_0+U_+} dU \int_{W_0}^{W_0+W_+} dW \cdot g. \tag{3}$$

Under tensile load there is an immense number of spatial ways for the intergrain cementation contact to be cleaved with the same basic result: the creation of crack. A similar multivariant scenario is true also for the already existing balanced crack to be further expanded. On the contrary, under compressive load the crack once emerged has only one spatial way in which to be annihilated or contracted. These observations are the principal ones and imply the huge disparity $\nu_0 \gg \mu_0$ between the priming rates ν_0 and μ_0 notwithstanding the generic cohesive properties of cementation material. Moreover, because of possible water intercalation and/or fragmentation of cementation material between the opposite faces of crack we can expect the typical value of barrier U to exceed that of barrier W. In combination all these factors might sustain even the more immense disparity $\nu \gg \mu$ between the actual rates ν and μ of defect creation and defect annihilation, apparently comprising many orders, and as a result provide the physical mechanism that breaks the symmetry of system response to an alternating external drive and acts as a sort of soft ratchet or leaky diode.

To express the evolution equation,

$$\rho\frac{\partial^2 u}{\partial t^2} = \frac{\partial \sigma}{\partial x} + \frac{\partial}{\partial x}\left[\frac{\partial \mathcal{F}}{\partial(\partial^2 u/\partial x\,\partial t)}\right], \tag{4}$$

for the field of longitudinal displacements u we choose the stress-strain relation in the form

$$\sigma = \frac{E\ \mathrm{sech}\ \eta}{(r-a)[\cosh\ \eta\,\partial u/\partial x + 1]^{a+1}} - \frac{E\ \mathrm{sech}\ \eta}{(r-a)[\cosh\ \eta\,\partial u/\partial x + 1]^{r+1}}, \tag{5}$$

which at $r>a>0$ allows one to block the bar compressibility at strain $\partial u/\partial x$ tending toward $+0-\mathrm{sech}\ \eta$. To put it differently, the parameter $\mathrm{sech}\ \eta$ is reserved for the typical thickness of intergrain cementation contact divided by the typical distance between the centers of neighboring grains. The dissipative function $\mathcal{F}$ we take in the form

$$\mathcal{F} = (\gamma/2)[\partial^2 u/\partial x\,\partial t]^2, \tag{6}$$

giving rise to Stokes internal friction [18]. Here x denotes the longitudinal Lagrange coordinate of the bar sample. The quantities ρ and γ are, respectively, the mean density of sandstone and the coefficient of internal friction in an elastic subsystem. We ignore their dependence on temperature and water saturation assuming that the main effect is manifested through the linear decrease of Young's modulus E with the concentration of defects,

$$E = (1 - c/c_{\mathrm{cr}})E_+. \tag{7}$$

Here c_{cr} and E_+ are the critical concentration of defects and the maximum possible value of the Young's modulus, respectively. Both of these parameters we also take to be independent of the temperature and water saturation.

Typical resonant response experiments [1–5] correspond to kinematic excitation [19] of a bar, which we associate with the following boundary conditions:

$$u(x=0|t) = D(t)\cos\left(\varphi + \int_0^t d\tau\omega(\tau)\right), \tag{8}$$

$$\frac{\partial u}{\partial x}(x=L|t) = 0, \tag{9}$$

where L is the sample length and $t>0$. The driving amplitude $D(t)$ is assumed to be basically constant except for the moments when the driving system is switched on, is switched into another constant driving level, or is switched off. The time dependence of cyclic driving frequency $\omega(t)$ in

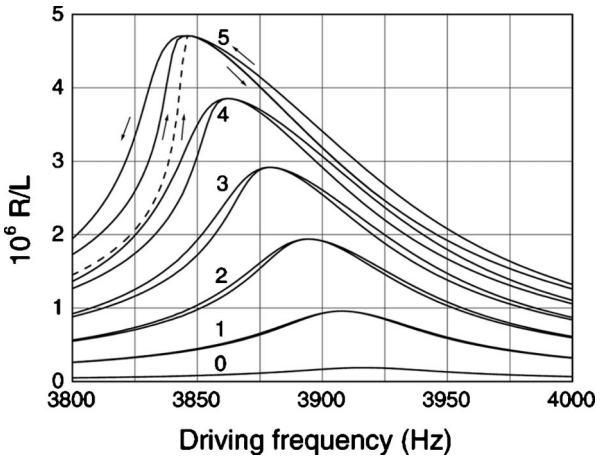

FIG. 1. Resonance curves $j=0,1,2,3,4,5$ at successively higher driving amplitudes $D_j=3.8(j+0.2\delta_{j0})10^{-8}L$. The time to sweep back and forth within the frequency interval 3700–4100 Hz is chosen to be 120 s.

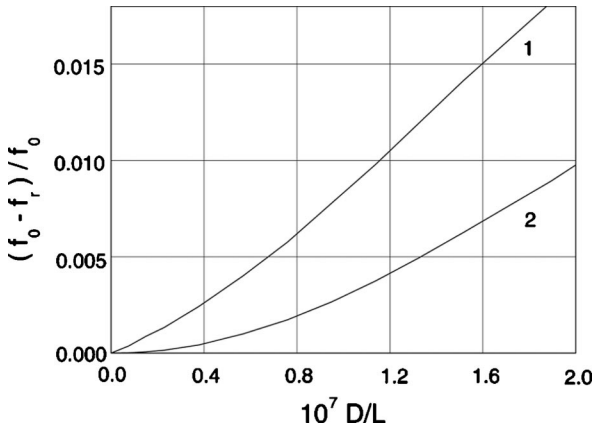

FIG. 2. Negative of the shift f_r-f_0 of resonant frequency f_r from its asymptotic value f_0 as a function of driving amplitude D for the hysteretic nonlinear material (curve 1) and for the classical nonlinear material with $v=0$ (curve 2).

turn is prescribed by the chosen regime of frequency sweep. Initial conditions are given in the form

$$u(x|t=0)=0, \quad \frac{\partial u}{\partial t}(x|t=0)=0 \quad (0<x<L), \quad (10)$$

$$g(x|t=0)=c_0/U_+W_+ \quad (0<x<L). \quad (11)$$

When experimental data for the Young's modulus in unstrained samples are obtainable from the resonant response experiments by the use of a low amplitude protocol (very small driving amplitude and negligible number of strain-induced defects), we can compare them with values taken from Eq. (7) at $c=c_0$ in order to fit the equilibrium concentration of defects c_0 as a function of T and s by some extrapolation formula. In particular, relying upon Sutherland temperature extrapolation [20] and analyzing temperature dependent data at zero saturation [21] and saturation dependent data at room temperature [13] for Berea sandstone, we suggest the formula

$$c_0=c_{\rm cr}\left(\frac{T}{T_{\rm cr}}\right)^2\left[\cosh^2\alpha-\exp\left(-\frac{\beta s}{1-s}\right)\sinh^2\alpha\right], \quad (12)$$

with the following fitting parameters: $T_{\rm cr}=1475$ K, $\cosh^2\alpha=16$, $\beta=10$. Here the saturation can vary within the interval $0\leqslant s\leqslant 1$. At $s\neq 0$ this approximation is expected to work at least for temperatures that exceed the freezing point of pore water.

Computer modeling of nonlinear and slow dynamics effects was performed in the vicinity of the resonant frequency $f_0(2)$, which we understand to be the second frequency ($l=2$) in the fundamental set,

$$f_0(l)=\frac{2l-1}{4L}\sqrt{\left(1-\frac{c_0}{c_{\rm cr}}\right)\frac{E_+}{\rho}} \quad (l=1,2,3,\ldots,), \quad (13)$$

given by the linear theory of kinematic excitation for zero dissipation $\gamma=0$.

Figure 1 shows typical resonance curves, i.e. dependences of response amplitude R (calculated at $x=L$) on drive frequency $f=\omega/2\pi$, at successively higher drive amplitudes D. The solid lines correspond to the conditioned resonance curves calculated after two frequency sweeps were performed at each driving level in order to achieve repeatable hysteretic curves. The dashed line illustrates an unconditioned curve obtained without any preliminary conditioning. Arrows on the three highest curves indicate sweep directions. For the sake of definiteness the results of the computer simulation were adapted to the experimental conditions supporting the data obtained by Ten Cate and Shankland for Berea sandstone [2]. In particular, the ratio E_+/ρ was estimated by means of relationships (13) and (12) with the second order frequency, bar length, temperature and saturation as follows: $f_0(2)=3920$ Hz, $L=0.3$ m, $T=297$ K and $s=0.25$. The ratio γ/ρ that characterizes internal friction was chosen from the best fit of the low amplitude theoretical curve (Fig. 1) to its experimental counterpart [2] using quality factor Q from the resonance width. The parameters $\mu_0\exp(-U_0/kT)=1$ s^{-1} and $U_+/k=2525$ K that determine the character of slow relaxation were estimated according to the experimental measurements of decay of acceleration at fixed frequency [2] and observations of recovering resonant frequency as a function of time [5]. The combination of parameters $vE_+/k\cosh\eta=275$ K was chosen to quantitatively reproduce the hysteretic phenomena in the sweep regimes typical for the actual experiments [2]. The nonlinearity parameter $\cosh\eta=2300$ was estimated to map the true asymmetry of experimental resonance curves [2]. Other parameters appearing in stress-strain relation (5) have been adopted as follows: $r=4$, $a=2$.

We would like to stress that through the drop of equilibrium concentration c_0 our theory is able to catch the dramatic suppression of hysteresis with decreases of water saturation [see Eq. (12)]. This conclusion has been confirmed by direct computation (not shown). Simultaneously we have observed a monotonic decrease in quality factor Q with an increase of saturation, i.e., precisely the well documented tendency in experiments [12]. In present theory this is due to the drop of resonant frequency with the water saturation [see Eq. (13)].

Figure 2 compares the shifts of resonant frequency as

functions of the driving amplitude at two essentially different values of dilatation parameter v while other parameters were kept the same as in Fig. 1. Thus curve 1 calculated at vE_+/k cosh $\eta=275$ K, when the strain-induced feedback between the slow and the fast subsystems is substantial, demonstrates the almost linear dependence typical of materials with nonclassical nonlinear response, i.e., materials that possess all the basic features of slow dynamics. On the other hand, curve 2 calculated at $v=0$, when the strain-induced excitation of the slow subsystem is absent and hence the mutual feedback between the slow and the fast subsystems is totally broken, demonstrates the almost quadratic dependence typical of materials with classical nonlinear response.

Finally, Fig. 3 shows the gradual recovery of resonant frequency f_r to its maximum limiting value f_0 after the bar has been subjected to high amplitude conditioning and conditioning is stopped. We have plotted three different curves corresponding to three different saturations with all other model parameters adopted earlier for Fig. 1 preserved. The total shift of resonant frequency f_r-f_0 consists of two physically different parts, namely, (i) the traditional dynamic shift caused by strain nonlinearity at high levels of excitation and (ii) the shift caused by the effect of the slow subsystem. However, only the second part might actually be registered during the recovery process, because the first one vanishes almost instantaneously when the conditioning was switched off. Hence, the whole character of recovery should inevitably be governed by the slow kinetics responsible for restoration of intergrain bonds. From Fig. 3 we clearly see the very wide time interval $10\leqslant(t-t_c)/t_0\leqslant1000$ of logarithmic recovery

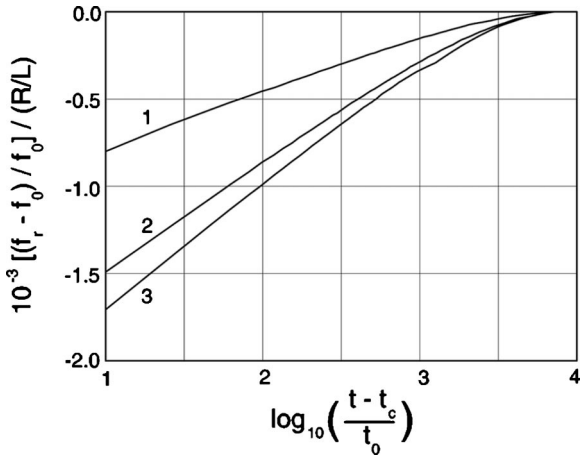

FIG. 3. Time-dependent recovery of resonant frequency f_r to its asymptotic value f_0. Curves $j=1,2,$ and 3 correspond to successively higher saturations $s_j=0.05(2j-1)$. The frequency shift f_r-f_0 is normalized by both the asymptotic frequency f_0 and the unitless response amplitude R/L attained at conditioning resonance.

of the resonant frequency, in complete agreement with experimental results [5]. Here t_c, is the moment when conditioning switches off and $t_0=1$ s.

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