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Tetsuro Suzuki
Missouri University of Science and Technology

Manfred Wuttig
Missouri University of Science and Technology

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Non-linear Oscillation, Automodulation and Anelasticity

Tetsuro SUZUKI and Manfred WUTTIG†

Institute of Applied Physics, University of Tsukuba, Sakura, Ibaraki 305
†Department of Metallurgican Engineering, University of Missouri-Rolla, Rolla, Missouri 65401, U.S.A.

The automodulation observed in the course of internal friction is discussed as a special example of the universal nature of non-linear oscillations. Universal features of non-linear oscillations are the requirement of energy supply or negative damping and the period doubling route to chaos. The nagative damping responsible for the automodulation is described in terms of the soliton model for twinning deformation.

§1. Introduction

Internal friction measurements have been established as a powerful tool to study the wide variety of properties of solids, including the atomic configurations of point defects and the tunneling states in crystalline and amorphous materials. However, there have been scattered reports of troubles in the course of the measurement of internal friction, which have sometimes been called "gasping". This denotes the situation where the amplitude of the oscillation in the course of the internal friction changes with an extremely low frequency compared with the oscillation frequency of the internal friction sample, despite the best efforts to keep the driving condition strictly stationary. The purpose of the present paper is to introduce this strange phenomenon of gasping or automodulation to the general audience interested in non-linear acoustic phenomena in various solids and liquids, and to try to establish the relationship between this special phenomenon and general non-linear oscillations in various fields of research.

§2. Non-linearity

The general definition of non-linearity is given in terms of the relationship between the input and the output of a system or black box. Suppose that we obtain the output $y_i(t)$ for the input $x_i(t)$, with $i=1$ or 2. If we obtain $\alpha y_1(t)+\beta y_2(t)$ as the output from the system for the input $\alpha x_1(t)+\beta x_2(t)$, the system is defined as linear.

If the output for the sinusoidal input is non-sinusoidal, the system cannot be linear and is considered non-linear. If a finite output is obtained for $\alpha=\beta=0$, i.e., for zero input, the system is called an oscillator. An oscillator can be considered as a special case of a non-linear system, even when the output waveform is sinusoidal. An oscillator cannot contradict the requirement from the first law of thermodynamics—the conservation of energy. A steady flow of energy is required for the steady oscillation to continue.

The non-linearity of a given system can be divided into two categories. Non-linearity (1) is that of the system into which no energy flow other than the input signal exists as shown in Fig. 1(a). This is the non-linearity generally expected from a system in thermodynamical equilibrium. Nonlinearity (2) is that of a system into which an energy flow other than the input signal does exist as shown in Fig. 1(b); this refers to the non-linearity shown by an oscillator. Because there is a flow of energy into the system, it represents the non-linearity of the system in thermodynamical non-equilibrium.

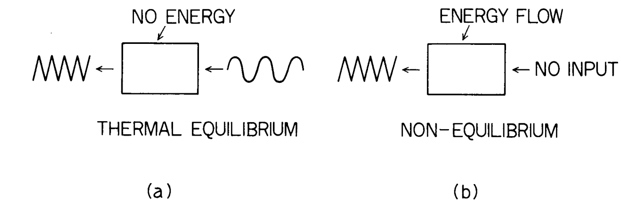

Fig. 1. The non-linearity (1) of the system in equilibrium (a), and the non-linearity (2) of the system in thermodynamic non-equilibrium (b).

2.1 *Non-linearity (1)*

This is the non-linearity shown by almost all kinds of material for sufficiently large input amplitudes. The deviation from Hooke's law $\sigma=C\varepsilon$ between the stress σ and the strain ε inevitably exists for any material with elastic modulus C. Hooke's law is the reflection of the parabolic dependence of the free energy of a material on the strain ε as shown in Fig. 2. Because any material can accommodate only a very limited value of the strain before it fractures or is plastically deformed, the parabolic dependence of the deformation free energy cannot possibly extend beyond a certain strain limit ε_c. In other words, this type of non-linearity (1) in elastic behavior is ubiquitous. The careful observation of this kind of nonlinearity by McSkimin and Andreatch[1] and Hiki and Granato[2] has lead to the

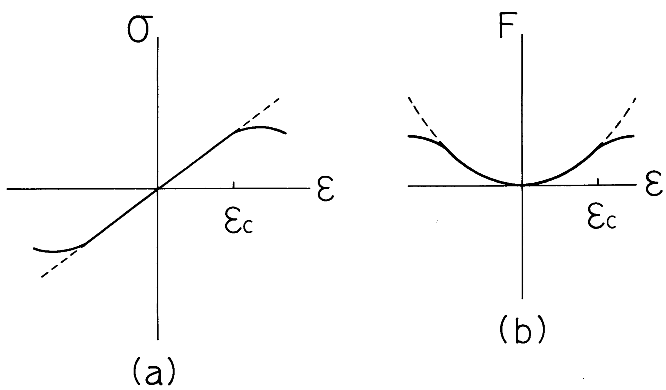

Fig. 2. The deviation from Hooke's law (a), and the nonparabolic dependence of free energy on the strain ε (b).

measurement of higher order elastic moduli. As a sophisticated utilization of this kind of non-linearity, the phonon echo experiment can be mentioned. However, the vast archives of non-linear acoustics deal with this kind of nonlinearity and cannot possibly be reviewed by the present authors in a way consistent with their importance.

2.2 *Non-linearity (2)*

Non-linearity (2) essentially deals with that shown by any kind of oscillator. A steady flow of energy is requried for positive feedback in the system to maintain the oscillation. A simple example of the non-linear oscillator is cited in Fig. 3 from the classical book by Stoker.[3] The mechanism for the music sound production in the violin has also been discussed by Rayleigh[4] in terms of the non-linear mechanism associated with the steady supply of energy. The bowing supplies the energy flow into the violin.

Although the steady flow of energy is a necessary condition for the oscillation, it is not by itself sufficient. Consider as another example of non-linear oscillation the pupulation variation in a colony consisting of several species of herbivorous (grass-feeding) and carnivorous (flesh-eating) animals described by the Volterra equations

$$\frac{dN_i}{dt}=\sum_j(\varepsilon_i-r_{ij}N_j)N_j, i=1,2,3,\ldots, \quad (2.1)$$

where N_i indicates the population of the i-th species of animals, ε_i the decay constant for the i-th species, and r_{ij} the constant indicating the predator-prey relationship between the i-th and j-th species. The decay constant ε_i for the herbivorous animals should be positive corresponding to the energy flow into the colony in the form of meadow grass for them, while that for the carnivorous animals should be negative. Equation (2.1) has stationary solutions given by

$$N_j=\varepsilon_i/r_{ij}. \quad (2.2)$$

It is possible that the population of different species takes on stationary equilibrium if the energy flow in the form of meadow grass maintains a sufficient population of the herbivorous species. If the population of one of the species grows larger than the stationary values given by eq. (2.2), oscillation in the population of the colony sets in. In other words, a certain level of energy supply into the colony in the form of meadow grass is necessary for oscillation in the population. The existence of a critical value for energy flow into the system seems to be a common feature of the oscillator in the system with non-linearity (2). The qualitative features of the trajectories of the solutions of eq. (2.1)

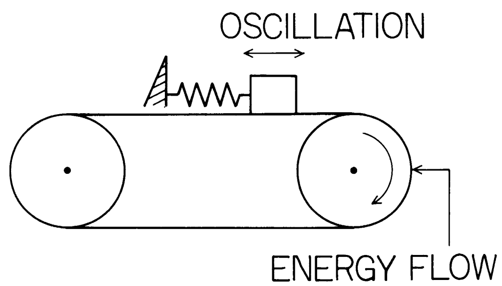

Fig. 3. Oscillation of a block on the belt by Stoker. The oscillation is sustained by turning the belt against the friction.

for increasing values of energy flow as studied numerically by Arnedo *et al.*[5] are shown schematically in Fig. 4.

§3. Bénard Convection and Oscillation

The Bénard convection in a fluid has been studied as a typical problem in the thermodynamical non-equilibrium state. The transitions from the regular convection pattern through successive transition patterns to turbulence have been studied by many authors.[6] When the temperature difference ΔT between the surface and the bottom of the fluid reaches a critical value ΔT_c, a mono-periodic regular convection pattern appears. In the Bénard convection experiment carried out by Maeno, Haucke and Wheatley[7] for the mixture fluid of $^4He+1.6$ mole%^3He, the critical temperature was 7.706 mK for the cell used in the experiment. At the temperature difference given by $\varepsilon\equiv\Delta T/\Delta T_c-1=3.77$, the regular steady periodic convection pattern set in. Figure 5 shows the observed power spectrum for oscillations at $\varepsilon=4.358$ and $\varepsilon=4.381$. The spectra clearly show the period doubling route to chaos (PDRC) proposed as a universal feature of non-linear oscillation by Feigenbaum.[8] The trajectory of the population described by the Volterra equation as shown in Fig. 4 is an ellipsoidal shape for small values of the energy input. But for increasing values of the energy input, a successive doubling of the trajectory period takes place as shown in Fig. 4.

§4. Automodulation in Internal Friction

The strain amplitude used in the internal friction is kept as low as possible, so that the non-linearity inevitably associated with large strain may be avoided. However,

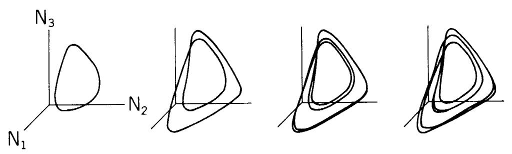

Fig. 4. Qualitative features of trajectories of the solution of the Volterra equation as studied by Arnedo, Coullet and Tresser.

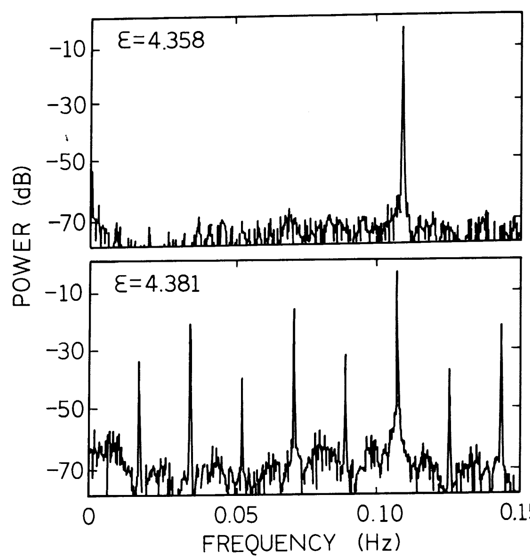

Fig. 5. The power spectrum of oscillation in a Bénard cell for $^4He+{}^3He$ mixture fluid.

when the amplitude is purposely kept large in the course of the measurement of internal friction and frequency, a strange slow modulation of the vibration amplitude has been reported by Mizubayashi and Okuda.[9] Even with the small strain amplitude for the internal friction, an inevitable instability of the amplitude of oscillation associated with the martensitic transformation in Cu–Al–Ni alloy has been reported by De Jonghe *et al.*[10] A similar instability associated with the twinning has been reported recently by Yokoyama.[11] These kinds of instabilities of the amplitude of the oscillation during the course of an internal friction measurement were also observed long ago by Takahashi[12] and Baxter and Wilks[13] and were referred to as "gasping".

A systematic effort to utilize this strange gasping or automodulation (of the amplitude of oscillation) for the study of the non-linear anelasticity associated with structural transformation has been initiated by Wuttig and coworkers.[14–17] The apparatus used to measure the automodulation is shown in Fig. 6. It is a standard apparatus to measure the internal friction by flexural vibration of the specimen, except the arrangement of Sm–Co magnets on the specimen and a pair of Helmholtz coils which allows the large amplitude sinusoidal excitation of the specimen. The left half of Fig. 7 shows the non-linear resonance curve of Mg single crystal specimen. As we shift the driving frequency from ν_4 to ν_1, the amplitude of the oscillation moves up along the non-linear resonance curve. The right half of Fig. 7 shows the variation of the amplitude measured as a function of time at four slightly different frequencies, hence

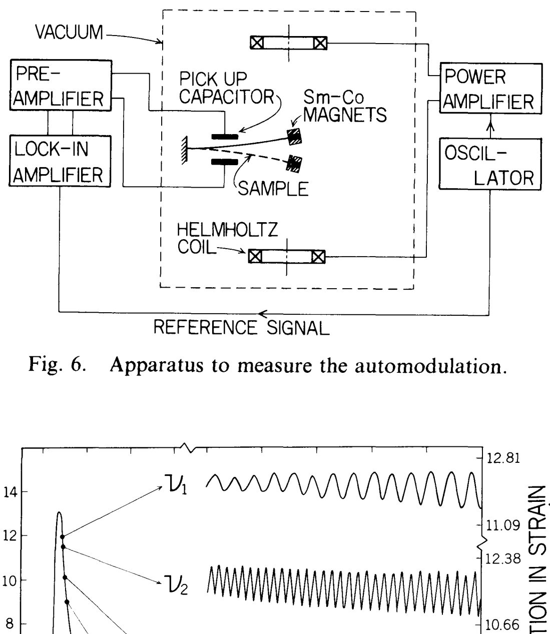

Fig. 6. Apparatus to measure the automodulation.

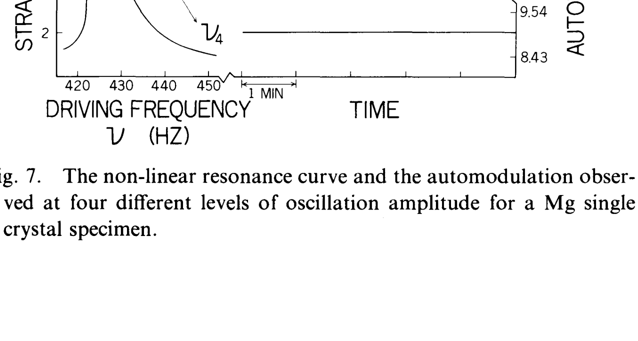

Fig. 7. The non-linear resonance curve and the automodulation observed at four different levels of oscillation amplitude for a Mg single crystal specimen.

at four different levels of oscillation amplitude. At the amplitude of oscillation larger than that corresponding to the driving frequency ν_3, a very slow automodulation of the amplitude is clearly seen. The automodulation frequency for the driving frequency ν_1 is doubled compared with the automodulation frequency for ν_3. A similar period doubling in the phase modulation pattern has also been observed. Furthermore, this type of period doubling, which is the universal feature of the non-linear oscillation, has been observed in the recent experiment on a In–Tl specimen above its martensitic transformation temperature.

§5. Anelasticity and Automodulation

Because the automodulation is observed when the amplitude of oscillation of the specimen for measurement of internal friction exceeds a critical value and the period doubling feature is also noticed, the automodulation may be interpreted as one variant of the oscillations of a nonlinear oscillator. The energy input for the oscillator is in the form of the driving of the specimen. The output is the modulation in the amplitude of the oscillation. Although the input energy and the output energy are in the same form, the input and output have entirely different frequencies. Hence, it is not entirely inappropriate to assign the name of non-linear oscillation to the automodulation phenomena.

The automodulation is shown to be expected[14] when internal friction measurement is carried out on a specimen which has the non-linear anelastic stress (τ) strain (ε) relationship

$$\tau + b\dot{\tau} = C(\varepsilon)\varepsilon + d(\varepsilon)\dot{\varepsilon}, \tag{5.1}$$

where b is a constant, and $C(\varepsilon)$ and $d(\varepsilon)$ are dependent on strain ε as follows:

$$C(\varepsilon) = C_2 + C_3\varepsilon + C_4\varepsilon^2 + \ldots, \tag{5.2}$$

and

$$d(\varepsilon) = d_2 + 2d_3\varepsilon + 3d_4\varepsilon^2 + \ldots. \tag{5.3}$$

While eq. (5.2) represents the stress-strain relationship in the completely relaxed state, eq. (5.3) divided by the constant b represents that relationship in the unrelaxed state, in other words, the response of the specimen to a suddenly applied stress or strain. The equation for the displacement u of a given point in the specimen for the fundamental mode of specimen vibration is given by

$$\rho\frac{\partial^2 u}{\partial t^2} + \left\{C_2 u + \frac{3}{4}C_4 u^3 + \ldots\right\} + \left\{d_2\frac{\partial u}{\partial t} + \frac{9}{4}d_4 u^2\frac{\partial u}{\partial t} + \ldots\right\} = 0, \tag{5.4}$$

neglecting the damping of external origin. This is a nonlinear differential equation with a non-linear damping represented by the terms in the second bracket, which has its origin in the non-linearity of the unrelaxed stress-strain relationship given by eq. (5.3). It is seen that if the nonlinear elastic modulus d_4/b in the unrelaxed stress-strain relationship is negative, the effective damping term can become negative for sufficiently large values of the amplitude of the specimen vibration amplitude, thus providing the mechanism for the automodulation. The characteristic of eq. (5.4) is that the negative damping, the maintenance of the automodulation, is possible only for finite values of the specimen vibration amplitude and the non-linear unrelaxed stress-strain relationship required for the negative damping.

§6. Anelasticity and Soliton Model

It has been discussed in the previous section that the non-linear characteristics of the unrelaxed modulus—the response of the specimen to a suddenly applied stress—is crucial for automodulation. In the In–Tl and Cu–Al–Ni alloys, in which the automodulation is observed, it is rather well established that the martensitic transformation in these alloys almost always accompanies the twinning process. Zn and Mg form another class of materials in which automodulation is observed. These metals are known to be deformed by twinning. The present authors propose that the automodulation comes from the non-linear anelasticity associated with twinning.

The standard procedure to study the nucleation process of the twin has been to use the classical nucleation theory, which assumes thermal equilibrium between nuclei and matrix. However, it is not appropriate to use the classical nucleation theory for the study of the dynamical process where the concept of the unrelaxed modulus is crucial. An entirely different approach can be taken by using the soliton model for structural phase transformation developed by Krumhansl and Schrieffer.[18] The soliton model has also been completed only for thermodynamical equilibrium. However, the effect of the suddenly applied stress can be more clearly seen by use of the soliton model than by use of the classical nucleation theory.

If we adopt the soliton model of Krumhansl and Schrieffer to the study of the twinning process, it is pictured as the coherent jumping of a chain of atoms—the motion of a soliton, as shown in Fig. 8. The atoms in the soliton model are situated in one of the double well potentials and neighboring atoms are coupled by the springs with a force constant κ. The atoms coupled by the springs are so similar to the linear chain model for the lattice that the role of the spring in the soliton model is quite likely to be misunderstood. The twinned and untwinned state of a real crystal is defined with respect to the relative configuration of atoms in the crystal, while the twinning process in the soliton model is pictured merely as the jumping of atoms over the barrier. Hence, the displacement in the model should be translated to the strain in the real lattice. Accordingly, the coupling spring in Fig. 8 should be referred to the energy associated with the gradient of the strain, i.e., the surface energy. The strain energy in the real crystal is represented as the potential energy of the double well in the model, but not as the coupling spring in Fig. 8.

As shown by the statistical theory for the soliton model, the formation of the soliton, i.e., the formation of the twin, is possible only below the temperature which is approximately determined by

$$\frac{(k_B T)^2}{\kappa a^2} \sim \text{(Barrier Energy between Double Well)}. \tag{6.1}$$

Here, a is the lattice constant and k_B the Boltzmann constant. Above the temperature T, the formation of the soliton—the formation of twin boundary—is overwhelmed by the random anharmonic vibration over the barrier. The role of the coupling spring in the soliton model or the surface energy in the real crystal is to organize the coherent motion of atoms for the formation of the soliton or twin boundary.

Suppose the external stress is applied adiabatically just below the temperature given by eq. (6.1), where the average amplitude of the vibration of the atoms in the soliton model is slightly lower than the height of the double well barrier. Then, the external stress creates a coherently overpopulated chain of atoms—analogous to the negative temperature distribution as shown in Fig. 9, which shows the statistical distribution[18] of atoms in the soliton model. The release of this overpopulated chain of atoms give rise to the negative non-linear unrelaxed modulus responsible for the automodulation.

§7. Summary

The non-linearity of a system—mainly a crystal or a liquid specimen—is classified into two types. The first type appears whenever a large enough input signal is applied to the system. The second type is that of an oscillator. Energy flow into the system or negative damping is necessary for an oscillator to function. As the amount of energy flow increases, the non-linear oscillation takes on the universal course, the period doubling route to chaos (PDRC), as proposed by Feigenbaum. As a typical example of PDRC, the non-linear oscillation associated with the Bénard convection experiment by Maeno, Haucke and Wheatley is cited. In the internal friction experiment, where the energy flow into the system is supplied in the form of the constant amplitude driving force of a specimen, the non-linear oscillation appears in the amplitude of the specimen vibration, which is called automodulation. This requires negative non-linear damping, which can be described as the release of the overpopulated states in the soliton model, for twinning.

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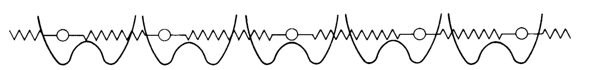

Fig. 8. Soliton model for the twinning. Each atom is situated individually in the double well potential and coupled to the neighboring atom by the coupling spring.

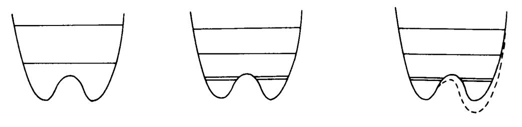

Fig. 9. The statistical distribution of atoms in the soliton model.

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