of the thermodynamic forces vanishing. This solution can be plausibly understood as a result of the normal counterflow driven by the magnetically changed chemical potential, Eq. (14), to eliminate the pressure difference $s\delta T+\rho\delta\mu$, and thus leading to the temperature drop $\delta T=-\delta\mu/\sigma$.

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Chaotic States of Anharmonic Systems in Periodic Fields

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It is shown that the nonlinear dynamics of anharmonically interacting particles in the presence of periodic fields leads to a set of cascading bifurcations into a chaotic state. This state is characterized by the existence of a strange attractor in phase space and associated broadband noise in the spectral density. It is suggested that solid-state turbulence is likely to be found in weakly pinned charge-density-wave systems and superionic conductors.

The problem of anharmonicity in condensed matter is an old and important one. Relevant to thermal expansion, heat conductivity, impurity modes, lattice dynamics,[1] and nonlinear optics,[2] to name a few topics, it led to the development of perturbation techniques that have been quite successful in explaining the experimental results. There exist a number of systems, however, for which small-amplitude results are not entirely appropriate. As exemplified by displacive phase transitions and superionic conductors, the dynamics of soft-mode behavior and ion transport is characterized by large atomic displacements which require nonperturbative theories. In the absence of damping processes, singular results have been obtained, indicating the existence of kinks or solitons.[3] These nonlinear solutions also appear as relevant degrees of freedom in field theory.[4,5]

In this paper we study the nonlinear dynamics of particles in anharmonic potentials in the presence of an external periodic field. As we will show, there exists a range of parameter values for which the solutions of the corresponding deterministic equation display a set of cascading bifurcations into a chaotic state, characterized by a strange attractor in phase space, and associated broadband noise in the spectral density. These features are reminiscent of the transition to turbulence encountered in stressed fluids,[5,6] some dissipative dynamical systems,[7] and simple mathematical models.[8] By constructing the return maps associated with the Poincaré sections of our problem we are able to determine that the transition to the chaotic regime belongs to the same universality class as the recursion relations recently studied by Feigenbaum.[9] We also determine the phase diagram in the amplitude-frequency plane and establish the existence of hysteresis effects. Finally, we suggest that solid-state turbulence is likely to be found in weakly pinned charge-density–wave (CDW) systems and superionic conductors.

Consider a particle of mass m and charge Q moving in a one-dimensional potential given by

$$V(\eta)=\tfrac{1}{2}a\eta^2-\tfrac{1}{4}b\eta^4, \qquad (1)$$

where η denotes the particle displacement from equilibrium, and the constants a and b are positive. Furthermore, we assume that the particle is subjected to a periodic external electric field of frequency ω.[10] If the coupling to all the other degrees of freedom of the solid is characterized by a phenomenological damping coefficient γ, the deterministic equation of motion for the charge reads

$$m\,d^2\eta/dt^2+\gamma d\eta/dt+m\omega_0^{\,2}\eta - b\eta^3-QE\cos(\omega t)=0 \qquad (2)$$

with $\omega_0^{\,2}=a/m$ and $\gamma>0$. In the limit of small fields and displacements, the solutions of Eq. (2) can be obtained via perturbation theory.[11] Within that scheme the effect of anharmonicity is to produce a renormalization of the damping coefficient and to cause mixing between the frequencies ω and ω_0. The phase portraits acquire some complexity but can, nevertheless, be analyzed in terms of simple limit cycles and fixed points. But as the amplitude of the electric field becomes large the consequent large-η behavior becomes increasingly difficult to treat perturbatively and the limit cycles encountered in the small-field limit become unstable. As we show below, in the large-amplitude regime one encounters a rich variety of behavior, which includes aperiodic solutions with very sensitive dependence on initial conditions.

In order to study Eq. (2), it is convenient to rewrite it in dimensionless form. Using the saddle-point solution, η_0, of the potential $V(\eta)$ as a natural length scale, and the inverse natural frequency ω_0^{-1} to set the basic time scale, we can rewrite Eq. (2) as

$$d^2\psi/d\tau^2+\alpha\,d\psi/d\tau+\psi-4\psi^3 = \Gamma\cos[(\omega/\omega_0)\tau] \qquad (3)$$

with $\psi=\eta/2\eta_0$, $\tau=t\omega_0$, $\alpha=\gamma/m\omega_0$, $\Gamma=\eta_0 QE/8V_0$, and where

$$V_0=a^2/4b \qquad (4a)$$

and

$$\eta_0=(a/b)^{1/2}. \qquad (4b)$$

Equation (3) was solved by using a Systron-Donner analog computer to obtain the phase portraits and time series for the spectral densities as a function of Γ and ω for fixed α. For values of Γ large enough so as to make the particle visit the anharmonic component of the potential ($|\psi|\sim\frac{1}{2}$), the bifurcation scheme that results differs markedly from that which one obtains using perturbation theory. In Fig. 1 we show the schematic phase diagram for fixed values of Γ and α as a function of the external field frequency, normalized to ω_0. Starting at point A, as the frequency decreases the solutions correspond to limit cycles of period 1 with increasing amplitude. Be-

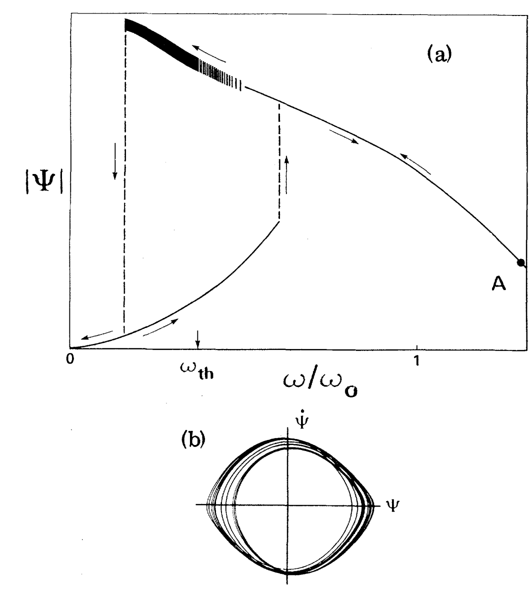

FIG. 1. (a) Schematic phase diagram for the anharmonic oscillator of Eq. (3). The thin solid lines denote periodic solutions, whereas the thick solid one corresponds to the chaotic state. The short vertical lines denote the set of cascading bifurcations. For the actual parameter values used ($\alpha=0.4$, $\Gamma=0.115$) we found $\omega_{th}\simeq 0.5567\omega_0$. (b) Phase portrait of the strange attractor for $\omega=0.552\omega_0$.

low $\omega/\omega_0 = 1$, a set of cascading bifurcations at frequencies ω_n starts taking place until a chaotic state, characterized by the appearance of a strange attractor in phase space [Fig. 1(b)], is reached at ω_{th}.[12] A more vivid picture of this transition can be obtained by looking at the power spectral density $S(\omega)$, which is depicted in Fig. 2. Each part of Fig. 2 is the average of ten spectral densities, obtained using the fast Fourier transform to process 4096-point time series which were initially shaped by a cosine bell window.

For values of $\omega > \omega_{th}$ [Fig. 2(a)] sharp peaks appear, which correspond to periodic states characterized by limit cycles of period $2^n T$, where T is the driving period. Beyond the chaotic threshold the power spectrum acquires the shape shown in Fig. 2(b). As can be seen, large-amplitude broadband noise appears, together with some well-defined frequencies. For lower values of ω [Fig. 2(c)] the system appears even more chaotic, and the strange attractor acquires a twofold band shape. If the frequency is lowered even further a sudden transition to an ordered state of period 1 takes place, a point below which perturbation theory becomes applicable. On reversing the sequence we have just described, the periodic behavior persists up to frequencies $\omega > \omega_{th}$, with a discontinuous jump into an ordered state with same period but larger amplitude. For our parameter values the hysteresis loop has $\Delta\omega = 0.143\omega_0$.

A sequence of Poincaré sections of the strange attractor, which we show in Fig. 2(d) at multiples of $\frac{1}{4}\pi$ in the driving phase, offers some insight into the nature of the chaotic state. As can be seen, it exhibits the typical folding process discussed by Shaw,[13] which signals the irreversible mixing and exponential divergence of trajectories. In fact, inspection of the sequence of sections reveals two complete folding processes per cycle of the driving force. For slightly larger driving frequency an asymmetric strange attractor, exhibiting a single folding process, was found.

In order to study in more detail the set of cascading bifurcations that precede the onset of the turbulent regime we constructed the return maps[8] associated with the Poincaré sections as $\omega \to \omega_{th}^+$ and for $\omega < \omega_{th}$. In this fashion we circumvented the errors associated with determining the exact frequencies at which new bifurcations set in. Although the details will be presented elsewhere, we observed that the return maps showed a single-hump feature with a parabolic maximum,

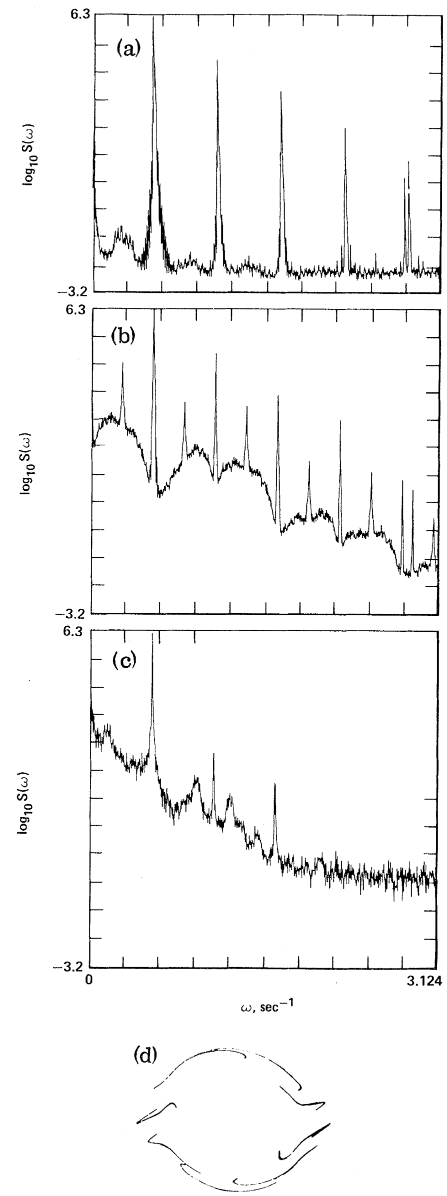

FIG. 2. (a)–(c) Fourier transform of the velocity autocorrelation function for several values of the field frequency with $\alpha = 0.4$ and $\Gamma = 0.115$, $\omega_0 = 1\ \text{sec}^{-1}$. (a) Driving frequency = 0.5623 sec^{-1}. (b) Driving frequency = 0.5558 sec^{-1}. (c) Driving frequency = 0.5529 sec^{-1}. (d) Poincaré sections of the strange attractor for $\omega = 0.5529\omega_0$ at multiples of $\frac{1}{4}\pi$ in the driving phase.

characteristic of the general class of recursion relations studied by Feigenbaum.[9] We are therefore able to ascertain that for large n the bifurcation sequence will display the same universal behavior, i.e.,

$$(T_{th} - T_n)/(T_{th} - T_{n+1}) = \delta, \tag{5}$$

where $\delta = 4.669\,201\,609\ldots$ and T_n is the value of the driving-force period at which the period-$2^n T_n$ limit cycle bifurcates to one of period $2^{n+1}T_{n+1}$.

The phenomena which we have just described could be found in solids whose anharmonic degrees of freedom can couple to a periodic field. Two likely candidates are weakly pinned charge-density waves (CDW) in anisotropic solids,[14] and superionic conductors.[15] In CDW systems, such as $K_2Pt(CN)_4Br_{0.3}\cdot 3H_2O$, tetrathiafulvalene-tetracyanoquinodimethane, and $NbSe_3$, it has been determined that charged impurities or commensurability act so as to prevent the CDW from freely sliding along the background lattice.[16-18] In that situation the enhanced electron density executes low-frequency (microwave to low infrared)[19,20] oscillations about the pinning center, and can be depinned by applying an external electric field of small magnitude (typically 10^{-9} V for $NbSe_3$). It should therefore be possible to drive the CDW in $NbSe_3$ into the turbulent regime by applying a microwave or infrared field of small magnitude in the temperature regime $59\,°K < T < 144\,°K$ (the region where only one CDW is present[21]. In fact, a recent experiment of Fleming and Grimes[22] reports conduction noise in $NbSe_3$ at temperatures below 59 °K, where the second instability sets in. Although this observation was made in the presence of a static electric field, it is possible that the coexistence of two CDW's might have produced a time-varying field of the type we have discussed.[23]

In superionic conductors, the ionic carriers oscillate around their potential minima with typically low optical-phonon frequencies[24] and hop over potential barriers with heights of order 0.1 to 0.2 eV. In particular, a class of superionic conductors displays large conduction anisotropies, a fact which renders the problem one-dimensional from our point of view. With $Li_2Ti_3O_7$ as an example,[25] it would require an infrared laser of moderate power to generate the phase diagram we have studied. The observation of the turbulent state could then be made by monitoring the absorption spectra.

In conclusion, we have shown that chaotic behavior is expected to occur in strongly anharmonic systems in the presence of periodic fields. Since the suggestions which we have made for the experimental search are by no means exhaustive, it is likely that such a phenomenon will be found in many other anharmonic condenced-matter systems.[26]

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