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Frequency locking for combustion synthesis in a periodic medium

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Abstract

Solutions of a 1D free-interface problem modeling solid combustion front propagating in combustible mixture with periodically varying concentration of reactant exhibit the classical phenomenon of mode locking. Numerical simulations show a variety of locked periodic, quasi-periodic and chaotic solutions.

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1. Introduction

The current Letter is intended to communicate new observations based on numerical solutions of the two-phase Stefan problem with kinetics. The simulations reveal some previously unknown features of this dynamically diverse system. Namely, it exhibits the phenomenon of frequency locking in response to a spatially-periodic perturbation of the medium.

The free-boundary problem that is the subject of the Letter arises naturally as a mathematical model of a variety of exothermic phase transition type processes, such as condensed-state combustion (also known as Self-propagating High-temperature Synthesis or SHS [7,9]), solidification with undercooling [4], laser induced evaporation [3], rapid crystallization in thin films [8], etc.

This study represents a natural extension of the numerical experiments described in [1], where it was demonstrated that, due to the competition between the heat release at the interface and the heat dissipation by the medium, the system generates a variety of complex thermokinetic oscillations. The dynamical patterns exhibited by the unperturbed system, as the governing parameters are varied, include Hopf bifurcation, period doubling cascades leading to chaotic pulsations, Shilnikov–Hopf bifurcation, etc.

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The new feature that was added to the setting is a variable initial concentration of the so-called deficient component which controls the reaction rate in the case of the combustion synthesis. We assume the concentration to be a periodically perturbed constant. While the original objective of our experiments was to study the dependence of the mean propagation velocity on the perturbation frequency and amplitude, we have noticed that the dynamics was qualitatively substantially different from that of the unperturbed problem. What we observed was a variety of quasi-periodic and complex periodic regimes with periods that are various multiples of the perturbation period. These observations present a convincing evidence that we are dealing here with the phenomenon of frequency locking.

The physical phenomena modeled by periodically driven dynamical systems appear in many fields (see, e.g., [10]) such as lasers, superconductors (Josephson's junctions), mechanical engineering, etc. Theoretical as well as numerical studies show that periodic forcing can drive these systems to exhibit rich patterns of behavior that includes mode locking, with the mode-locked bands usually having the structure of the so-called Arnold's tongues in the amplitude–frequency parameter space.

It is necessary to mention that the analysis or numerical simulation or a combination of thereof is usually carried out for a finite-dimensional system even if the original physical system is infinite-dimensional. The reduction in such cases is based on an assumption of a certain ansatz which consequently leads to an ODE. At the same time the perturbation is usually external to the base system and it is periodic in time. In our case, however, we make no attempt of such reduction, neither are we aware of any ansatz for a solution leading to it, while *the internal forcing is periodic in space*, and the system remains formally autonomous.

Below we present some examples of numerical solutions and a crude map of a basic resonance band in the amplitude–frequency parameter space that represents an Arnold's tongue. We remind the reader that every point in the parameter space is a result of numerical simulation of a non-trivial free-interface problem for a partial differential equation.

Thus, the sharp interface model of combustion synthesis presents a very natural and transparent example of the frequency locking phenomenon for a PDE.

2. Free-interface problem

In the context of combustion of condensed matter a wave of exothermic chemical reaction transforms a solid combustible mixture directly into solid product. The 1D model involves differential equations for the temperature u of the mixture and the relative concentration of the so-called deficient reactant Z (see, e.g., Shkadinsky et al. [11]):

$$u_t = \kappa u_{xx} + qW(Z,u),$$
$$Z_t = -W(Z,u),$$

where κ is the thermal diffusivity, W is the chemical reaction rate, and q is the heat release.

Due to a strong temperature dependence of the reaction there is a well-defined narrow region (flame front) where the bulk of chemical reaction and the heat release occur. Thus the distributed chemical reaction can be replaced by the δ-function (see Zeldovich et al. [12]),

$$W = Z\big(s(t)\big)g(u)\delta\big(x - s(t)\big),$$

located at the interface $x = s(t)$ between the fresh, $Z = Z(s(t))$ and burned $Z = 0$ material.

The equation with the δ-function source is rewritten as a system of two heat equations coupled at the interface. In the context of solidification with undercooling [4] or rapid crystallization of thin films [8], the free-interface model studied below is conceptually even simpler: the latent heat of the phase transition released at the interface must be diffused by the surrounding matter.

Therefore we shall be concerned with the following *appropriately non-dimensionalized* free-interface problem: find $s(t)$ and $u(x,t)$ such that

$$u_t = u_{xx}, \quad x \neq s(t), \tag{2.1}$$
$$u(x,0) = u_0(x) \geqslant 0, \tag{2.2}$$
$$Z_0\big(s(t)\big)g\big[u\big(s(t),t\big)\big] = v(t) \quad \text{for } t > 0, \tag{2.3}$$
$$u_x^+\big(s(t),t\big) - u_x^-\big(s(t),t\big) = v(t) \quad \text{for } t > 0, \tag{2.4}$$

where $v(t)$ is the interface velocity, $s(t) = \int_0^t v(\tau)\,d\tau$ is its position, u is the temperature, and the derivatives u_x^+ and u_x^- are taken from right side and left side of the free interface, respectively. At $-\infty$ the surrounding matter is assumed to be at the temperature of the fresh combustible, while the temperature of the burned matter as well as its gradient is bounded:

$$u(-\infty,t)=0, \qquad u(\infty,t)<C,$$
$$\left|u_x(\infty,t)\right|<C. \tag{2.5}$$

Under reasonable assumptions on the kinetics functions g, one can rigorously prove that the free-interface problem (2.1)–(2.4) possesses global in time, uniformly bounded classical solutions (see [2]). One should not regard the rigorous proof of the existence and uniform boundedness of solutions as a futile academic exercise but rather view it as a verification of correctness of the model. It provides a firm foundation for the numerical simulations presented in the Letter. In particular, the proof of uniform boundedness of solutions underscores the dynamical robustness of the model.

3. Overview of dynamics for unforced problem

For reader's convenience in this section we give a brief overview of the unforced dynamics (see [1]), i.e., with $Z_0(x)\equiv 1$. It is convenient to rewrite the non-equilibrium interface condition (2.3) in the form:

$$v=g\big[u\big(s(t),t\big)\big]:=1+\alpha J\big(u\big(s(t),t\big)\big). \tag{3.1}$$

We shall assume that the function $J(\xi)=(g(\xi)-1)/\alpha$ is normalized in such a way that

$$J(1)=0, \qquad J'(1)=-1, \tag{3.2}$$

which can be achieved by rescaling variables. This assumption makes the problems with different kinetics identical in terms of linearization about the basic solution. We note that the variables are selected so that the *interface propagates to the left*.

In order to clarify the meaning of the (positive) parameter α which is the main instability parameter, we note that for the Arrhenius type kinetics

$$v=g(u)=-\exp\left[\frac{\alpha(u-1)}{\sigma+(1-\sigma)u}\right], \tag{3.3}$$

α is the scaled activation energy for the exothermic chemical reaction that occurs at the interface, and σ is the temperature ratio of the fresh and burned material for the traveling wave solution.

The problem (2.1)–(2.4) has a unique traveling wave solution

$$u_b=\begin{cases}\exp(x+t), & x\leqslant -t,\\ 1, & x>-t,\end{cases} \qquad s_b=-t, \tag{3.4}$$

provided J is monotone. The linear stability analysis indicates that the loss of stability occurs via a supercritical Hopf bifurcation at $\alpha_{\rm cr}=\sqrt{5}+2$, the corresponding frequency is $\omega_{\rm cr}\simeq 1.03$, $T_{\rm cr}=2\pi/\omega_{\rm cr}\simeq 6.1$.

We have demonstrated in [1] that depending on the parameters, the system exhibits dynamical regimes that include a Hopf bifurcation followed by classical Feigenbaum's cascade of period doubling leading to chaotic pulsations, a Shilnikov–Hopf bifurcation, etc.

For numerical simulations below we employed an explicit finite-difference numerical code in the coordinate system attached to the interface $\eta=x-s(t)$. Problem (2.1)–(2.4) is considered on a finite interval $-L\leqslant\eta\leqslant L$ with the Dirichlet condition $u(-L,t)=0$ simulating the decay of the solution at $-\infty$ and Neumann condition $\frac{\partial\theta}{\partial\eta}(L,t)=0$ reflecting stabilization of the temperature far behind the interface in the burned matter (solid phase). In view of the fact that the dynamics of the problem requires a very fine temporal resolution (to resolve sharp changes in the velocity), our experience shows that there is no advantage in using implicit methods in this case (see [1] for detail).

We note that in mathematical terms, the free boundary problem in (2.1)–(2.4) governs the temporal evolution in the infinitely-dimensional "phase" space of functions $u(x)$ and scalars v. One way to present results would be through graphs of $u(x,t)$ versus x, t and time histories $v(t)$ vs. t. We also represent dynamics through projections of the infinitely-dimensional phase space onto the 3-dimensional space $[u(s(t)-1,t), u(s(t),t), u(s(t)+1,t)]$. I.e., the functional profile $u(\cdot,t)$ is represented by three values: at the interface and at two points equidistant from it.

4. Frequency locking

The main objective of this Letter is to demonstrate the phenomenon of frequency locking for the driven free-boundary problem (2.1)–(2.4). To this end we choose the initial mass concentration to be in the form

Table 1
Arnold's tongue of resonances (▽) in the (ω, a)-plane; $0.5 \leqslant \omega \leqslant 1.5$ (horizontally), $0 \leqslant a \leqslant 0.1$ (vertically)

▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽
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	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽	▽
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of the harmonically perturbed unity:

$$Z_0(s) = 1 + a\cos(\omega s). \tag{4.1}$$

Note that unlike typical forced problems, the perturbation here is a function of the dependent variable of the problem $s(t)$. Thus, strictly speaking, the problem remains autonomous. Therefore the driving frequency ω and the frequency of the periodic solution, as seen in the power spectra and temporal dynamics in the figures below, are not the same. Of course, it is possible to introduce a new time $\tau = s(t)$, which would make the perturbation "time"-dependent and the problem non-autonomous. However, it introduces a stiff highly-variable coefficient $1/v$ at the time derivative of the heat equation that makes numerical solution substantially more challenging.

Parameters in (3.3) were selected to be $\sigma = 0.1$, $\alpha = 4.5$ which correspond to a regime with simple periodic oscillations just past the Hopf bifurcation for the unperturbed problem. We vary the forcing amplitude a from $a = 0$ to $a = 0.1$ with an increment of 0.005 and the frequency ω from $\omega = 0.5$ to $\omega = 1.5$ with the step $\Delta\omega = 0.05$. The results show typical structures of the Arnold's tongues in the (a, ω)-parameter plane. Note that for every pair of parameters the computation involves numerical solving a non-trivial free-interface problem, and the computational cost of an exhausting analysis of an extended domain in the parameter plane becomes prohibitive.

Table 1 represents a crude outline of one of Arnold's tongues. We note that in addition to the main tongue one can also observe what seems to be smaller resonant tongues that correspond to higher order resonances. The resolution of the computations however, does not allow us to make a definitive judgement concerning additional tongues.

Next we discuss several representative solutions of the forced problem for various parameter sets. Each of the figures below contains a velocity profile (top), its power spectrum (bottom-left), and a 3-dimensional projection of the orbit into the space $[u(s(t) - 1, t), u(s(t), t), u(s(t) + 1, t)]$, where $u(\cdot, t)$ is the temperature (bottom-right).

Fig. 1 depicts a simple periodic orbit. Fig. 2 shows a trajectory whose period corresponds to twice the period of the forcing (note that the forcing has a *spatial* not temporal period). Figs. 3 and 4 depict periodic orbits of high multiplicity in ultraharmonic and subharmonic domains, respectively. Fig. 5 demonstrates an example of what appears to be a chaotic trajectory, which can be incurred from the power spectrum. This might indicate a presence of a period-doubling cas-

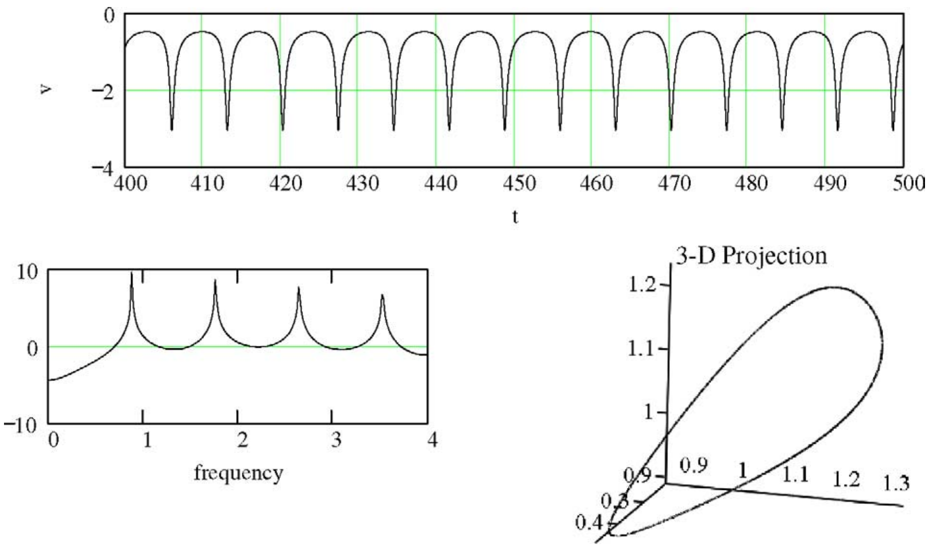

Fig. 1. $a = 0.04$, $\omega = 1$. Top: velocity profile; bottom-left: power spectrum; bottom-right: 3D projection of the orbit.

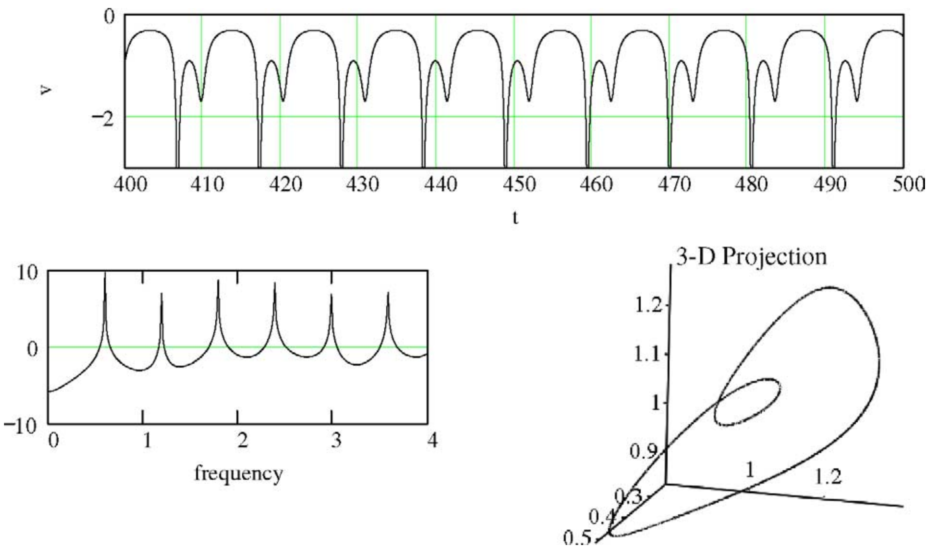

Fig. 2. $a = 0.08$, $\omega = 1.4$.

cade. Finally, Fig. 6 shows a typical quasi-periodic orbit. We remark that quasi-periodic solutions with more than two basic periods have also been observed.

5. Concluding remarks

Above we have demonstrated that the free-boundary problem (2.1)–(2.4) exhibits some basic behavior quite analogous to the prototypical finite-dimensional frequency locking systems such as, for instance, the driven van der Pol equation. Since the sole objective of this Letter is to demonstrate the presence of the phenomenon of mode locking for the free-interface problem we have deliberately chosen a very limited setting, and restrained ourselves from discussing a multitude of other questions that arise in the context of this complex dynamical phenomenon.

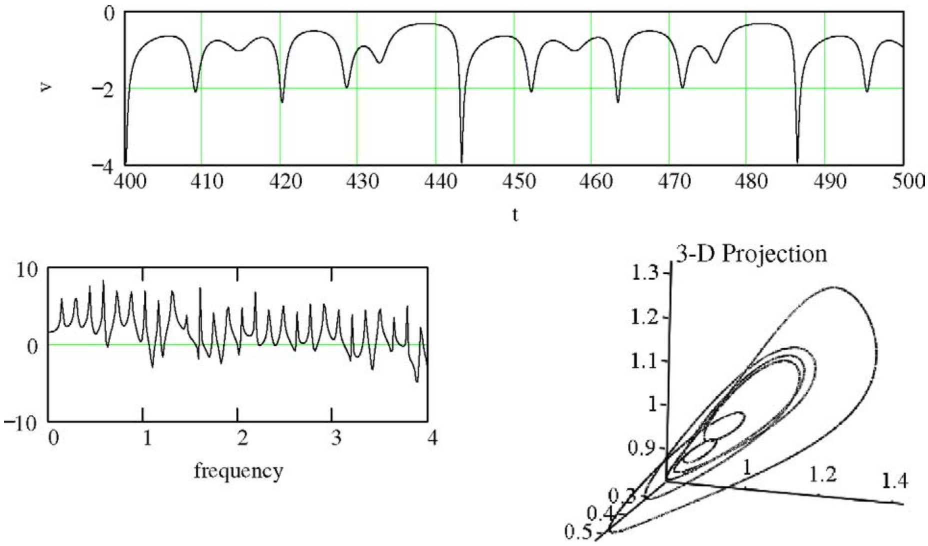

Fig. 3. Ultraharmonic resonance of multiplicity 4 for $a = 0.08$, $\omega = 0.65$.

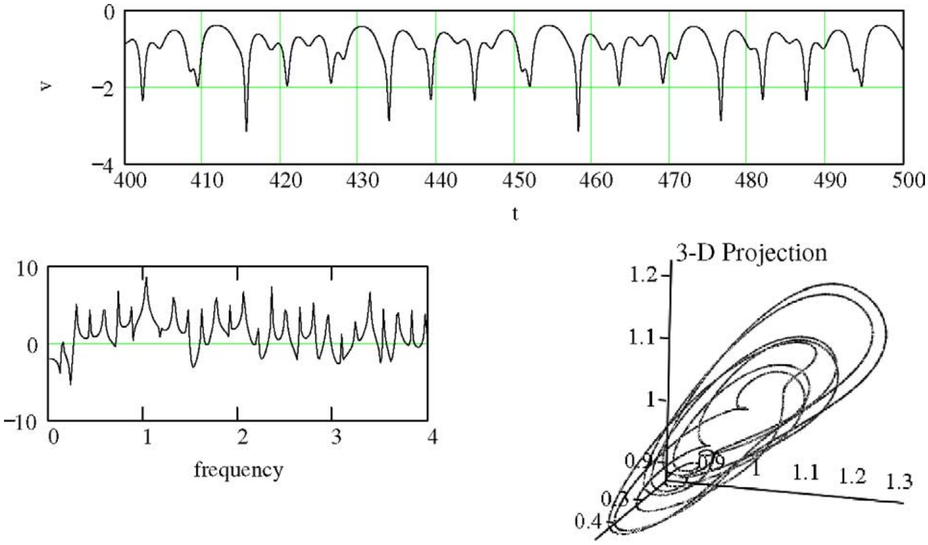

Fig. 4. Subharmonic resonance of large multiplicity for $a = 0.08$, $\omega = 2.5$.

There are a number of complex features observed in dynamical response of the van der Pol system [5,6] and other finite-dimensional counterparts that one would attempt to verify for our system. One would naturally ask, for instance, whether the Arnold's tongues represent a dense set with its complement forming a Cantor set in the (a, ω)-plane, whether the Feigenbaum sequences occur within the tongues, etc. It should be noted that the dynamics of the unforced free-boundary problem varies dramatically as the non-linearity parameter α increases. In this Letter we considered only the simplest possible case corresponding to stable limit cycle following Hopf bifurcation. It would be extremely interesting to investigate how the fre-

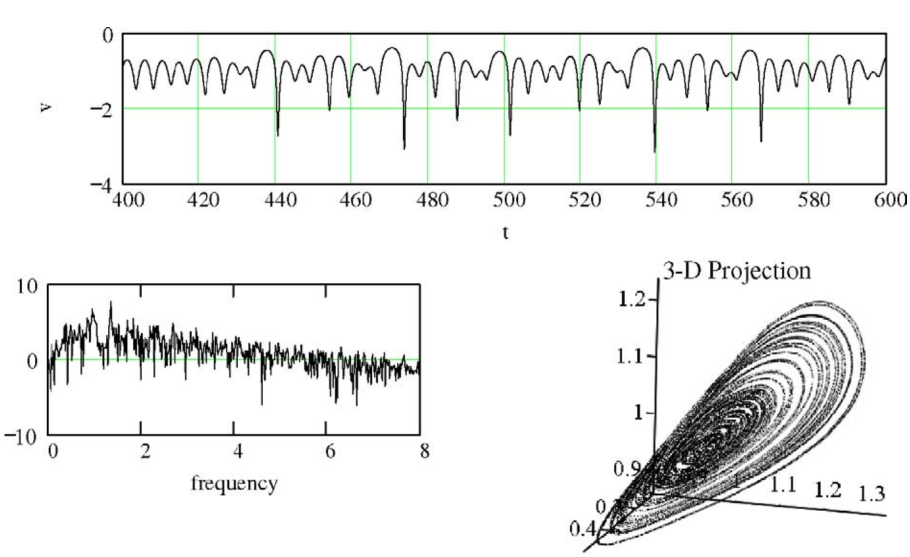

Fig. 5. Chaotic orbit for $a = 0.04$, $\omega = 1.4$.

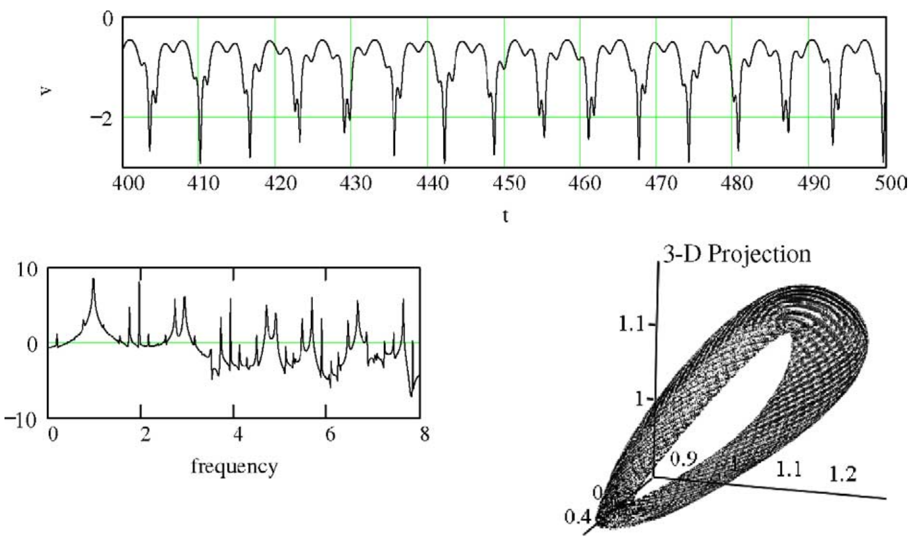

Fig. 6. Typical quasi-periodic orbit, $a = 0.1$, $\omega = 4$.

quency locking response changes with increasing α. We hope to be able to pursue these issues in the near future.

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