Nonlinear natural engine: Model for thermodynamic processes in mesoscale systems

(thermoacoustics/solitons/self-focusing/heat pumping)

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ABSTRACT To develop intuition on the possible application of concepts from thermodynamic heat engines to mesoscale systems, we have constructed and studied a model thermoacoustic heat engine. The model consists of a chain of coupled nonlinear acoustic vibrators in which the primary thermodynamic medium is argon gas, the secondary thermodynamic medium is constituted by solids bounding the gas, and frequencies are $ca. 3 \times 10^2$ Hz. The nonlinear elements are the necks, made flexible by means of an oil-loaded DuPont Kapton film, of Helmholtz resonators. When the primary medium is driven uniformly by an acoustic driver at a frequency somewhat below the low-amplitude resonant frequency and at a high enough driving amplitude, stationary localized or solitary states appear irreversibly on the chain. These are characterized by a higher vibrational amplitude than that in surrounding vibrators, where the amplitude can decrease; by the appearance of deep subharmonics of the drive frequency, corresponding to driven low-frequency vibrations of the Kapton film–oil systems; and by the pumping of heat toward the localized states. Possible implications of these results for mesoscale systems consisting of chains of molecular vibrators are then discussed.

The qualities of intrinsically irreversible acoustic heat engines (1, 2), or “natural” engines* for short, are so general that one might expect to find many examples of them in nature, not only very large and of low frequency, as in some variable stars (3), but also on a mesoscale,† intermediate between atomic and macroscopic sizes, and operating at very high frequencies. As a first step in studying the possible existence of mesoscale engines we have constructed and studied experimentally a thermoacoustic model, a nonlinear natural engine, consisting of a chain of coupled discrete nonlinear vibrators where thermodynamic symmetry (1, 2) is broken dynamically, under novel and suggestive circumstances, and a heat-pumping effect is observed. The choice of the properties of the model was motivated both by the work of Scott and colleagues (4) on Davydov-type (5) solitary waves in peptide chains at 5×10^{13} Hz and by the work of Wu *et al.* (6) and of Larraza and Putterman (7) on a hydrodynamic soliton at 5 Hz in a trough of water. In the usual natural acoustic engine (1, 2) heat pumping takes place along a stack of plates (a second medium) of limited spatial extent and in imperfect thermal contact with a fluid (a first medium), vibrating with an attendant oscillatory temperature change (thermodynamic effect). The thermodynamic symmetry between the first and second medium is broken at the ends of the stack; one end cools while the other heats, heat being pumped toward the pressure antinode for a fluid of positive expansion coefficient. The same sort of effect would also take place for an extended second medium, if the region of oscillatory motion and oscillatory temperature change in the

first medium were confined spatially. As this localization of vibratory motion can occur in a system of coupled nonlinear vibrators (4–7) at a sufficiently high level of excitation, we say that in this case the thermodynamic symmetry is broken dynamically, rather than geometrically as with the usual stack of plates. In a molecular system, such a localized state might serve as the mesoscale object that we would ultimately like to study.

The thermoacoustic model was built to resemble a molecular chain and was driven to produce a localized from an extended state. The nonlinearity involved “coupling to low frequency modes,” a terminology suggested by Scott *et al.* (8). We discovered that the dynamics of this coupling is very important and that deep, not necessarily commensurate, subharmonics of the drive are responsible for and attend both the formation of a localized state and the heat pumping. Such driven subharmonics may be essential in producing heat engine effects in mesoscale systems. The arrangement of the model is shown schematically and in plan view in Fig. 1 *Upper*. Twelve equal acoustical vibrators, or Helmholtz resonators, each consisting of two equal bulbs of volume V connected by a neck of area A and length ℓ , are arranged in a ring, the inner bulbs of the vibrators being interconnected azimuthally by tubes. An acoustic driver is located in the center. It is coupled to each of the bulbs by 12 equal moderately high flow impedance tubes, with the intent at low amplitude of driving a uniform extended oscillation of the vibrators in the ring. Individual sensors allow the dynamic pressure to be measured at the driver and at each of the 12 inner bulbs. The neck of each vibrator is a nonlinear element (for details see ref. 9). Important details are shown in the sectional drawings of Fig. 1 *Lower*. The gas in each neck occupies a space 11 mm wide by 3.6 mm high by 76 mm long; the upper half is bounded by a heat-formed DuPont Kapton film 0.025 mm thick and loaded with oil. The exits from neck to bulbs are flared to reduce turbulence. A high impedance tube connects the inside of each inner bulb to the surroundings so that the mean pressure p_m is the same inside and out. Design of the neck was based on the following. When the vibrator is excited the pressure in the center of the neck is less than the mean pressure by the Bernoulli pressure $\rho v^2/2$, where ρ is gas density and v is the acoustic velocity; so under vibrator excitation the film moves inward according to its elasticity, and the area of the neck decreases. The angular resonance frequency ω_0 of one vibrator is given by $\omega_0^2 = 2c^2(A/\ell V)$, where c is the sound velocity. Hence ω_0 decreases as the amplitude of vibration increases. With this characteristic for ω_0 , an analysis of a lossless ring when extended to the continuous limit, and following Larraza and Putterman (7),

Abbreviation: TAC, thermoacoustic couple.

*The adjective *natural* refers to the natural irreversible processes that are essential to the phasing between motion and thermodynamic effect in such engines.

†We are indebted to Eschel Ben-Jacob for introducing us to this descriptive term.

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FIG. 1. (Upper) Schematic plan view of a nonlinear acoustic model used to drive localized vibrational states and to observe the attendant heat-pumping effects. (Lower) Transverse and partial longitudinal sections of the neck of a typical acoustic vibrator.

yields a nonlinear Schrödinger equation for the bulb pressure amplitude, suggesting that in this limit a localized state like the hydrodynamic soliton would be observed if the ring were driven sufficiently hard at an angular frequency below the low-amplitude value of ω_0 .

Heat pumping effects should occur in the coupling tubes between the inner bulbs when vibrations in adjacent bulbs are not equal. To observe this effect we inserted in each coupling tube a thermoacoustic couple (TAC) (2) consisting of a single thin sheet of G-10 fiberglass with an electrical thermopile having five chromel-constantan thermocouples at each end. The heat pumping is toward the region of higher dynamic pressure and is given by the formula

$$|\dot{Q}| = \frac{1}{4} \Pi \delta_\kappa \frac{1 + \sqrt{\sigma}}{1 + \sigma} p_1 v_1, \quad [1]$$

where Π is the perimeter of the surface along which heat is being pumped, δ_κ is the thermal penetration depth, σ is Prandtl's number, and p_1 and v_1 are the amplitudes at the TAC of the dynamic pressure and velocity, presumed in

quadrature. In a coupling tube we have $v_1 = \Delta p_1 / \omega \rho \ell_N$, where Δp_1 is the amplitude of the pressure difference between the bulbs adjacent to the TAC, ω is angular frequency, and ℓ_N is the effective length of the coupling tube. The heat pumping sensed by a TAC is taken to be representative of the acoustically stimulated heat flow along the walls of the coupling tubes.

Each bulb volume V is 33.4 cm^3 , neck dimensions are given above, the coupling tubes are 5.2 mm in diameter by 41 mm long, and the coupled vibrators, characterized by $Q \approx 20$, resonate at low amplitude at *ca.* 283 Hz when filled with argon gas at room temperature and at $p_m = 4 \text{ bar}$ (1 bar = 10^5 Pa). Measurements of resonance curves using helium-4 gas, and thus near 900 Hz, are characteristic of the classical nonhysteretic one-dimensional nonlinear vibrator (10). But at the lower frequencies with argon gas the response of a single vibrator can be hysteretic for a drive frequency and amplitude where, based on the measurements with ^4He and nonlinear vibrator theory (10), the displacement of the Kapton film owing to the average Bernoulli pressure differ-

ence could not produce hysteresis. Moreover, with argon the frequency spectrum of the bulb pressure shows the presence of both commensurate and incommensurate subharmonics of the drive frequency. Deep subharmonics, corresponding frequently to frequency divisions by factors greater than 10, appear as sidebands of the central drive frequency. They appear as the drive is increased and are particularly strong after a transition has occurred to a hysteretic state, corresponding to a discontinuity in the average geometry of the neck. Visual observation under stroboscopic light and in air of the principal low-frequency mode of the Kapton film-oil system attending the transition to a hysteretic state shows that it is primarily a transversely symmetric vibration with a *ca.* half a wavelength across the membrane and with a shift in phase of π on passing longitudinally through the center of the neck. Rough calculations based on the theory of vibrations in plates (11) suggest that this is an elastic wave involving both the Kapton's elasticity and the oil's mass taking place at approximately constant total gas volume. Very-low-frequency modes are also observed, very likely a consequence of gravity waves on the oil. The existence of deep subharmonics is well known (12, 13) in arrangements not dissimilar from ours. The hysteretic character of the vibrator response at only moderately high dynamic pressure is then explained by a high-amplitude modulation of the neck geometry resonantly driven at the subharmonic frequency, the amplitude being much larger than the displacement expected from average Bernoulli pressure alone.

Some rather typical results of increasing the relative drive amplitude P_{oD}/p_m , where P_{oD} is the amplitude of dynamic pressure at the driver, are shown in Fig. 2 for argon gas at $p_m = 4.15$ bar, room temperature, and a drive frequency of 275 Hz, about 8 Hz below the low-amplitude resonance frequency. Here are plotted quantities $\delta p_n \equiv [(P_{oD}/(P_{oD})_F) - 1]$ and $\delta(TAC)_n \equiv (TAC)_n - [(TAC)_n]_F$ vs. azimuthal position. The quantities P_{oD} and $(TAC)_n$ are, respectively, the dynamic pressure amplitude at the n th inner bulb and the electromotive force generated by the n th TAC, while $(P_{oD})_F$ and $[(TAC)_n]_F$ are these quantities measured at a reference value of P_{oD}/p_m , 0.0252 in this illustration. This method of representing the data emphasizes the principal effect but fails to show the systematic differences from vibrator to vibrator actually observed and reflecting not only our inability to fabricate all the necks the same but also possibly small differences in the impedances coupling the ring to the driver. At the reference relative drive of 0.0252, vibrator 6 had the highest amplitude, some 6% above its neighbors 5 and 7 and some 11% above vibrators 11 and 2, which had the lowest amplitudes. $(TAC)_n$ is located adjacent to vibrator n and counterclockwise from it as indicated in Fig. 1. The polarity of $(TAC)_n$ is such that a positive electromotive force indicates that the end of $(TAC)_n$ adjacent to bulb n is hotter than its other end. Referring to Fig. 2, we see that at a relative drive of 0.0234 both δp and $\delta(TAC)$ show only minor changes from their values for the reference drive. However, at a somewhat higher relative drive, just below 0.0269, the ring makes an irreversible transition to a condition in which there is a localized state of higher vibrational intensity centered on bulb 11. (Just below this transition, scaling from the ^4He measurements, the drive was sufficient, via average Bernoulli pressure, to reduce the resonance frequency by only 3 Hz.) $(TAC)_{11}$ has a positive output while $(TAC)_{10}$ is negative, showing that heat is being pumped toward bulb 11 from both sides. On increasing the drive somewhat further a new localized state appears on bulb 5 attended by heat pumping of the correct sign while the state near bulb 11 broadens. It is interesting to note that the relative pressure amplitudes in bulbs between the localized states actually diminish, reflecting the "focusing" of the drive energy into the localized states. The sign and relative magnitudes of the heat pumping

FIG. 2. Incremental reduced dynamic pressures δp (●) in the various bulbs and the changes in the outputs of the thermoacoustic couples $\delta(TAC)$ (■) in the various coupling tubes plotted against azimuthal position for several values, increasing from top to bottom, of the reduced dynamic pressure amplitude P_{oD}/p_m of the driver. Both δp and $\delta(TAC)$ are defined in the text. The reference state is defined to have $P_{oD}/p_m = 0.0252$. The numbering of the azimuthal bulb and TAC positions is as in Fig. 1. The acoustic medium is argon gas at 4.15 bar. The drive frequency is 275 Hz, 8 Hz below the low-amplitude resonant frequency.

effect, as reflected in the TAC outputs, are in accord with the variations in δp . At higher drive yet the localized states broaden and shift, and eventually there is nearly equal excitation of all the vibrators. Excepting relatively small amplitude changes, the localized states do not move along the ring as long as P_{oD}/p_m is held constant.

For the illustration of Fig. 2, at $P_{oD}/p_m = 0.0252$ the strongest sideband of the dynamic pressure in bulb 11 within 50 Hz of the drive was displaced 4.6 Hz; at 0.0269 this sideband was gone and a stronger one displaced 25.6 Hz had developed. When bulb 5 makes its transition at 0.0279, sidebands develop displaced 25.9 Hz. Principal sideband displacements of the localized states experimentally need not be the same, though differences are only 1 or 2%. Within the precision of the measurements, of which Fig. 2 is representative, at other drive frequencies between 265 and 280 Hz, the sideband displacements have a tendency to shift with the drive frequencies with the ratio of drive to principal sideband displacements varying in the range 10.5 to 11.5 for the localized states. The principal sidebands are observed in all the vibrators but are largest at the localized states. In other qualities of the same series of measurements the dynamic pressure of vibrator 11 just below the transition to a localized state increases by a factor of *ca.* 1.4 as the drive frequency increases from 265 Hz to 280 Hz, respectively 18 Hz and 3 Hz

below the low-amplitude resonance frequency; it is relatively easier to excite a localized state at lower frequencies.

In other experiments with only two coupled vibrators, 9 and 10, all others having been removed, the general characteristics with respect to hysteretic instabilities, sidebands, heat pumping between vibrators, and drive frequency qualities were essentially the same as for the 12-vibrator arrangement of Fig. 1. For the drive frequency just below the low-amplitude resonance frequency of 287.5 Hz, large-amplitude subharmonics were observed. An example is shown in Fig. 3 for a drive frequency of 283.5 Hz and a full horizontal sweep of 2 sec. The dynamic pressure in bulb 9 is shown in the upper trace, modulated by a 48:1 subharmonic. The lower signal is the heavily filtered and amplified output of the TAC between inner bulbs 9 and 10, showing a modulated heat flow between the bulbs at the deep subharmonic frequency. For the very-low-frequency modes reflected in these measurements one usually finds oscillations that on the average are incommensurate with the drive but show a sudden phase slippage every few cycles.

These experiments show that the first localized states to form as drive is increased are centered on only one or two vibrators. The localization process is irreversible and dependent subtly on the properties of individual vibrators. While the attendant heat pumping cannot be calculated accurately from the TAC outputs, owing to the uncertain return heat path with each TAC tightly coupled to the walls of its coupling tube, the heat pumping effect expected is qualitatively confirmed. The average Bernoulli pressure difference is quantitatively inadequate to explain the hysteretic change observed and fails completely to account for the observation that as the drive frequency is displaced farther below the low-amplitude resonance frequency the dynamic pressure at the transition decreases. Yet it is a quantity analogous to the average Bernoulli pressure's effect on the vibrator geometry that enters theories (14) describing the self-focusing of waves via equations related to the nonlinear Schrödinger equation.

What is necessary to produce the irreversible transition in a given vibrator appears to be a *dynamic* "coupling to low frequency modes" (8), probably a resonant excitation of a deep and not necessarily commensurate subharmonic of the drive. With respect to localization, this is the most important and suggestive observation in the present work.

These experiments are helpful in developing intuition to study the mesoscale heat pumping problem. The molecular vibrators whose vibrational intensity is localized may well exist in long hydrogen-bonded chains and have frequencies of order 10^{12} Hz, corresponding to librational motions (15, 16). This frequency is probably too high for any heat pumping effect. However, localization may be facilitated and detected by coupling to a longitudinal subharmonic of the librational frequency, the physics of driving the subharmonic being perhaps similar to that in our model. It is known that longitudinal waves on DNA are not strongly damped if their frequency is high enough (17), and such a density wave could in principle be trapped in the region of conformational change attending the localized state. The angular frequency of the subharmonic may be estimated by $\omega = c/\ell$, where c is the longitudinal sound velocity and ℓ is the size of the localized state. For $c = 10^5$ cm/sec and $\ell = 100$ Å, $\omega = 10^{11}$ sec $^{-1}$. Heat pumping might be expected from this subharmonic longitudinal motion owing to irreversible thermal contact with the surroundings of the chain (the second medium), provided that the heat-carrying excitation mean free path, $3\kappa/c$, is much less than the classical thermal penetration depth, $(2\kappa/\omega)^{1/2}$. In that case a space- and time-dependent temperature distribution can exist, as would be necessary for a thermodynamic engine effect. For a thermal diffusivity κ of 10^{-3} cm 2 /sec and other values of the parameters given above one has $3\kappa/c \approx 3$ Å and $(2\kappa/\omega)^{1/2} \approx 14$ Å, so the necessary conditions for the second medium would be roughly fulfilled. It is also essential that an oscillatory adiabatic temperature be definable in the primary medium; the source of this thermodynamic effect is the effective linear expansion coefficient, probably negative,

FIG. 3. Oscilloscope photograph illustrating (upper trace) a 48:1 subharmonic modulation of the dynamic pressure of one of two coupled acoustic vibrators excited at high amplitude and at 283.5 Hz, 4 Hz below the low-amplitude resonant frequency. The amplified and filtered output of the thermoacoustic couple located in the coupling tube between the two vibrators is shown in the lower trace, illustrating a pulsating heat flow at the subharmonic frequency. The full sweep shown is 2 sec.

arising from the coupling of longitudinal and librational motions. Evidence for a negative linear expansion coefficient along the direction of the chain of hydrogen-bonded molecular vibrators has been reported by Forss (18) for L-alanine crystals.

In recent work Lomdahl and Kerr (19) have concluded from finite-temperature molecular dynamics calculations that the Davydov soliton (5) suggested to occur on hydrogen-bonded peptide chains with one quantum of amide-I energy will not have a significant lifetime near 300 K. One can conclude from their work that this soliton is a useful excitation only if the thermal energy $k_B T$, where k_B is Boltzmann's constant and T is absolute temperature, is substantially less than the self-trapping energy. This result seems to rule out an important role for molecular-sized localized states if longitudinal motion is important. However, we conjecture that the mesoscale structures suggested here may be almost semiclassical objects excited to a very high vibrational level with self-trapping energies larger than $k_B T$ near room temperature. In that case both a mesoscale region of high vibrational intensity and a concomitant trapped density wave of much lower frequency may exist to perform a useful function. In spite of the highly speculative nature of these conjectures, they should be useful in motivating further experimental work.

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