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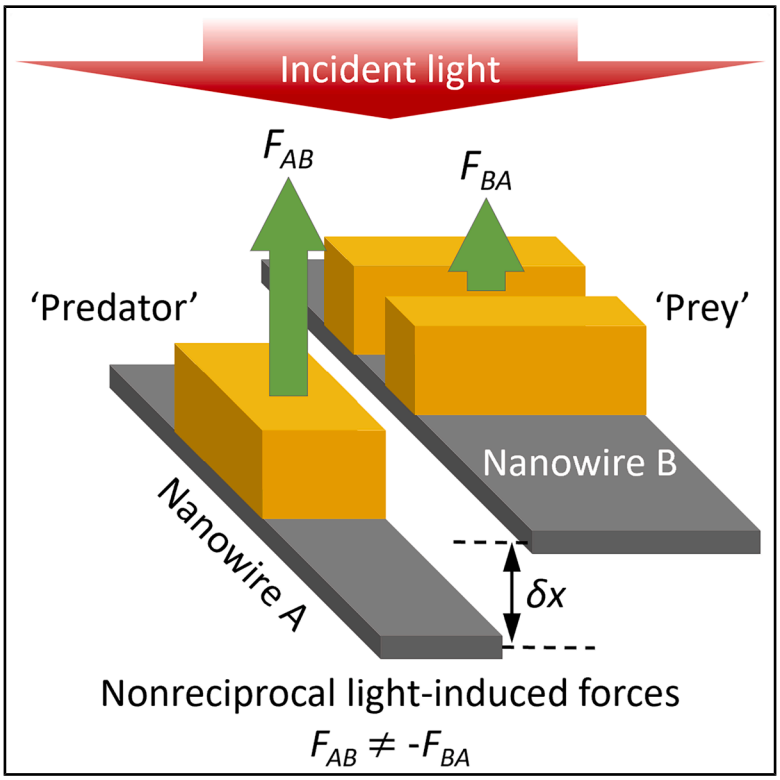
Time-translation symmetry, ergodicity, and entropy dynamics in a time crystal driven by optical interaction forces

Graphical abstract



Highlights

- Nanomechanical oscillator ensemble driven to time-crystal state by light
- Transition triggered by nonreciprocal optical interactions between oscillators
- Time-translation symmetry and ergodicity broken; entropy locally decreased
- A material platform for "timetronics"—data processing based on time crystals

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In brief

There is growing interest in non-equilibrium active matter, which converts a source of energy into motion. Liu et al. study the dynamics of an ensemble of nanoscale oscillators and show how light can induce nonreciprocal interactions between oscillators, triggering persistent coherent oscillations across the ensemble. The breaking of ergodicity and time-translation symmetry signifies a transition to the time-crystal state, which is of interest for implementing timetronics—a new data processing paradigm based on time crystals.

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Time-translation symmetry, ergodicity, and entropy dynamics in a time crystal driven by optical interaction forces

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ACCESSIBLE OVERVIEW There is growing recognition that nonreciprocal interactions play an important role in the dynamics of biological populations and in colloid chemistry. Here, we show that light can induce nonreciprocal interactions between nanoscale mechanical oscillators, triggering a transition to the time-crystal state, spontaneously breaking ergodicity and time-translation symmetry, and locally lowering entropy—characteristics that are also manifested in living matter. The ability to engineer such behaviors in artificial materials presents exciting opportunities for developing "timetronics"—a new data processing technology based on time crystals—and for studying the dynamics of many-body systems.

SUMMARY

Nonreciprocal processes are universally present in living systems, and they control social behavior, kinetic asymmetries in chemistry, pattern formation, and phase transitions in active matter out of equilibrium. It was shown recently that light-induced "predator-prey" interactions in an ensemble of pairs of dissimilar nanoscale oscillators can facilitate the transition to the continuous time-crystal state of matter. Here, we experimentally study the dynamics of time-crystal transitions driven by nonreciprocal optical interaction forces. We observe that the breaking of continuous time-translation symmetry and ergodicity and a decrease in entropy characterize such transitions. Intriguingly, such dynamics are also manifested by living matter. We argue that nonreciprocally driven time crystals represent a powerful platform for studying the classical dynamics of many-body systems, including modeling the matter-to-life transition, and present an opportunity to implement all-optical neural networks and reservoir computing architectures.

INTRODUCTION

Nonreciprocal processes in systems out of equilibrium are attracting growing interest across the entire domain of scientific research,[1,2] from sociology to nanotechnology, including a recent suggestion that nonreciprocal interactions may have played a critical role in the origin of life, i.e., the matter-to-life transition.[3] Such interactions are ubiquitous in living systems and underpin the behavior of inhomogeneous social groups, swarming, and hierarchical and predator-prey dynamics.[4–6] They also arise naturally in molecular systems,[3,7] for example, due to diffusion and kinetic asymmetries as in "predator-prey chasing" among oil droplets of different chemistries, and they lead to time-dependent phases as illustrated by the dynamics of interacting autonomous vehicles.[2] In many-body ensembles, nonreciprocal interactions initiate symmetry breaking and lead to pattern formation, hysteresis, and phase transitions.[2,6,8] In open systems, light can induce nonreciprocal interaction forces due to differential scattering, for example, between suspended (optically trapped/bound) nanospheres[9–11] or the constituent nanostructural elements of photonic metamaterial arrays.[12,13] Indeed, it has recently been demonstrated that such interactions facilitate the realization of the continuous time-crystal state of matter in a plasmonic nanowire metamaterial.[14,15]

A time crystal, first discussed by Frank Wilczek in 2012, is a many-body interacting system that exhibits a spontaneous

transition to a robust oscillatory state, breaking time-translation symmetry under an arbitrarily small change in the external driving force.[16,17] Two types of time crystals are subsequently defined: "discrete" time crystals break discrete time-translation symmetry by oscillating at a sub-harmonic frequency of periodic external driving force and "continuous" time crystals break continuous time-translation symmetry by transitioning to robust periodic motion (with arbitrary phase) in response to a time-invariant force.[18]

Various realizations of time crystals using spins, Bose-Einstein condensates, magnons, the qubits of a quantum computer, and other exotic nonlinear systems have been demonstrated.[19–22] However, the continuous time crystal realized in Liu et al.[14] is uniquely driven by nonreciprocal optical interaction forces[10,13,23] that arise within an array of linear nanowire oscillators, alternately decorated with dissimilar plasmonic nanoparticles: above a threshold level of light intensity, the nonreciprocal interaction forces trigger a spontaneous transition in the nanowire ensemble, from a state of uncorrelated stochastic thermal motion to a state of persistent, synchronized, high-amplitude periodic oscillation.[15] Here and below, following Zheludev,[24] such transitions and the reverse transition back to the thermal state will be referred to as "mobilization" and "demobilization," respectively.

In a system where an electromagnetic field is present, nonreciprocal interaction forces between dissimilar objects can emerge from the radiation pressure induced by scattered fields.[9] For example, intense scattering from a larger object "B" will act upon a smaller object "A" with a force F_{AB} that may be much stronger than the oppositely directed force F_{BA} associated with weaker scattering from the smaller object acting on the larger. Thus, $F_{BA} \neq -F_{AB}$, breaking action-reaction equality and therefore interaction reciprocity. The nonreciprocal component of interaction can be significant in nanoscale systems. Indeed, nearfield plasmonic effects and Mie scattering, dependent on the size, shape, refractive index, and separation of nanoparticles, can resonantly enhance interaction forces[12,13] and thus departures from the action-reaction equality, even to the extent that interaction forces become co-directed.[15] It should be noted here that violation of the action-reaction equality by objects interacting in the presence of light does not contradict Newton's third law, which applies only to isolated systems.

The transient dynamics of light-induced mobilization and demobilization in this nonreciprocity-driven continuous time crystal have not previously been studied. In this work, by monitoring transitions in the nanowire ensemble in the time domain, we experimentally obtain the mobilization and demobilization times of the time-crystal state. We provide the first experimental study of ergodicity breaking in a time crystal by mapping the phase of the demobilized, mobilized, and intermediate transient states, and we demonstrate that the entropy of the nanowires' motion is locally reduced upon mobilization. We also observe that the phase of mobilized oscillation is random with respect to the triggering event, confirming that continuous time-translation symmetry is spontaneously broken upon mobilization.

Intriguingly, the above characteristics of the optically stimulated time-crystal transition—reduction of entropy and spontaneous breaking of time-translation symmetry and ergodicity—are also key characteristics of living matter, in which light is often the main source of energy.

RESULTS

Experimental study of mobilization and demobilization dynamics

We studied a metamaterial array comprising alternately dissimilar dielectric nanowires decorated with plasmonic nanorods, fabricated from a gold-coated silicon nitride membrane (see methods), such that the nanorods form split Π-shaped metamolecules, each supported on a pair of neighboring nanowires, as illustrated on the left-hand side of Figure 1. Mutual displacement of the nanowires reconfigures the metamolecules, changing their plasmonic resonances and thereby the optical properties[23,25]—the transmissivity, reflectivity, and absorption—of the array, providing a means for detecting its state optically. Metamolecule dimensions are chosen such that light at a wavelength λ = 1.55 μm, close to the structure's plasmonic absorption resonance, induces nonreciprocal out-of-plane interaction forces between the two parts of the unit cell supported on neighboring nanowires[15]: in the terminology of nonreciprocal dynamics,[7] the nanowire supporting a single gold nanorod (of the three in each metamolecule) becomes a "predator" chasing the "prey"—i.e., the other nanowire supporting the two-nanorod part of the metamolecule—which in turn tries to move away from the predator. The strength of this nonreciprocal interaction increases with light intensity and, upon reaching a threshold value, triggers spontaneous synchronization of nanowire (oscillator) movement across the array. (The experimentally observed time-crystal state is based upon the out-of-plane flexural mode,[14,15] as in-plane optical forces do not provide predator-prey coupling between the two parts of the metamolecules.)

Below the synchronization threshold, the nanowires exhibit independent stochastic thermomechanical movement driven by momentum transfer from flexural phonon creation and annihilation. According to the Langevin theory of thermally driven oscillators, these fluctuations are most profound at the natural frequency of the oscillator. In the present metamaterial array, the resonance frequencies ω_{0i} of the individual nanowires' out-of-plane motion lie around $2\pi \times$ 877 kHz (with some dispersion due to fabrication imperfections and inhomogeneity in the membrane's intrinsic stress). At room temperature, the nanowires fluctuate independently with time-averaged displacement amplitudes $\langle x_T \rangle = \sqrt{\frac{k_B T}{m_i \omega_{0i}^2}}$ of order 300 pm at their mechanical resonance frequencies (where m_i is the nanowire effective mass, T is temperature, and k_B is the Boltzmann constant). These picometric fluctuations are already sufficient to measurably change the reflectivity and transmissivity of the array,[25] and upon mobilization, neighboring nanowires begin to move in antiphase, their amplitudes of oscillation increase by up to two orders of magnitude (increasing the amplitude of transmissivity changes), and the frequency spectrum of their collective motion collapses.[14,15]

We observe in experiments that the array exhibits a spontaneous mobilization transition to the synchronized oscillatory state at a threshold laser power P_M = 39 μW (intensity

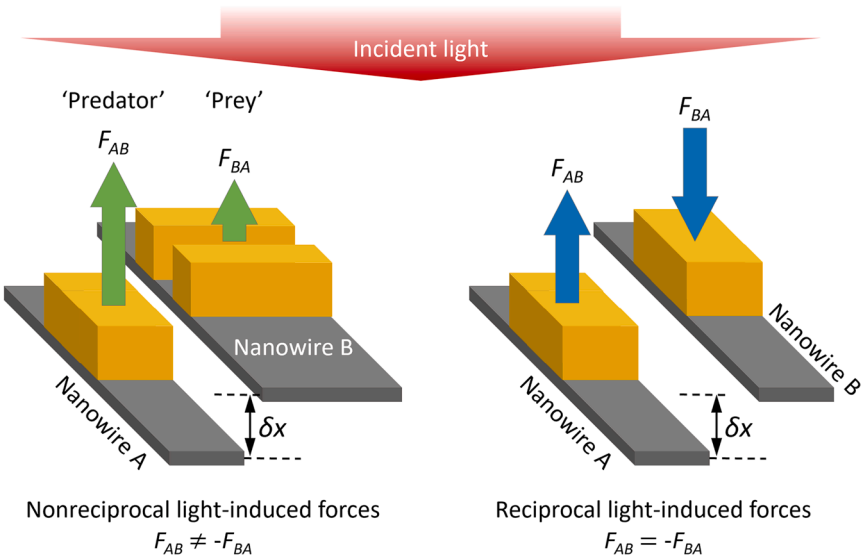

Figure 1. Predator-prey dynamics induced by light in a nanowire plasmonic metamaterial

When there is mutual out-of-plane displacement δx between neighboring dissimilar nanowires, light scattered on the supported plasmonic nanorods asymmetrically changes the balance of optical forces acting on the wires. If the additional forces are nonreciprocal (asymmetric, as illustrated on the left) $F_{AB} \neq -F_{BA}$, a predator-prey dynamic can be established, and above a threshold of light intensity, nanowire oscillations will synchronize in antiphase. If the additional forces are reciprocal $F_{AB} = -F_{BA}$, i.e., equal in magnitude and opposite in direction (as would be the case between identical nanowires, as illustrated on the right), the motion of the nanowires remains unsynchronized. Whether the interaction forces are reciprocal or nonreciprocal depends on the geometry of the metamolecules and wavelength of incident light.

~2.0 μm/μm^2 over a full-width half-maximum spot diameter of 5 μm; see methods). With decreasing power, the demobilization transition occurs at P_D = 36 μW. In what follows, we explore the dynamics of these transitions by applying sharp square-wave step changes in incident laser power across the thresholds (about a constant average power $P_{avg.}$ = 29.4 μW, as shown by the blue traces in Figures 2A–2C) using an electro-optic modulator. We record time-series data of the photodiode current and then filter the signal to retain only frequency components in the range Δf from 845 to 885 kHz, encompassing the natural resonance frequencies of all illuminated nanowires' out-of-plane motion. By normalizing this signal against the zero-frequency component of the photodiode current, which is proportional to sample transmissivity τ_0 at $P_{avg.}$, we arrive at normalized transmissivity change $\delta\tau(t)/\tau_0$—a parameter that depends directly on the state of the nanowire array ($\delta\tau(t)$ being transmissivity change in the spectral range Δf associated with the out-of-plane oscillation of the nanowires).

While incident laser power remains at all times below the P_M = 39 μW synchronization threshold, as in Figure 2A (where it is switched between two sub-threshold values of ~21 and ~37 μW) the normalized transmissivity change fluctuates about zero with low amplitude, due to the incoherent thermal motion of the nanowires and detection system noise.

Figure 2B shows transmissivity dynamics at the onset of synchronization, when incident laser power is increased abruptly from ~17 to ~40 μW, which is to say from markedly below to just above threshold. While the power is above threshold, the amplitude of transmissivity modulation slowly increases (mobilization). The rise time is unstable from one synchronization cycle to the next, varying from ~20 to ~35 ms (from ~17,000 to ~30,000 oscillation cycles). When the incident peak power is further above threshold (~43 μW in Figure 2C), the synchronization dynamics stabilize—the mobilization transition occurs faster, with a rise time reduced to 15 ms. In contrast, the decay/relaxation time of the reverse (demobilization) transition, from the synchronized state to the incoherent (thermally fluctuating) state, does not depend on incident laser power.

In Figures 2D–2F, we plot the instantaneous spectral entropy of transmissivity modulation, which is a measure of the disorder of oscillations, or randomness of the system. It is defined as $H = -\sum_{m=1}^{N} P(f_m)\log_2 P(f_m)$, where the probability distribution $P(f_m) = \frac{S(f_m)}{\sum_i S(f_i)}$, and $S(f_m)$ is the power spectrum of the process. The entropy decreases slowly from the onset of mobilization and increases rapidly at the demobilization transition. These data show that the mobilized state is characterized by a stable entropy level below that of the disordered state.

With an abrupt decrease in incident laser power from above to markedly below the mobilization threshold, the oscillators revert to the regime of unsynchronized, incoherent thermomechanical fluctuation. The amplitude of oscillation decays exponentially with a relaxation, or demobilization, time $1/\gamma$ (γ being the damping parameter), observed to be ~40 μs (~35 cycles, Figure 3A). The oscillators also dephase at a rate of $\pi/(2\delta\omega_0)$, where $2\delta\omega_0$ is the inhomogeneous broadening of the array, which is longer than the relaxation time, leading to a complex demobilization process.

To further reveal the dynamics of mobilization, we plot transmissivity phase diagrams ($\delta\tau$ against $\partial\delta\tau/\partial t$) for different stages of the transition (Figures 3C–3E). In the disordered state, i.e., while incident laser power is below the mobilization threshold, characteristically stochastic motion is observed with a Gaussian phase distribution (Figure 3C). When incident power is increased above the threshold, there is a transition (Figure 3D) to a limit cycle of stable synchronized motion (the mobilized state, Figure 3E).

Boltzmann's ergodic hypothesis postulates that the trajectory of an isolated mechanical system runs through all states of the system, which in the case of the demobilized system are formed by the modes of individual nanowires. In the presence of light, the isolated status of the system is removed, and upon a transition to the mobilized state, the phase trajectory of the system

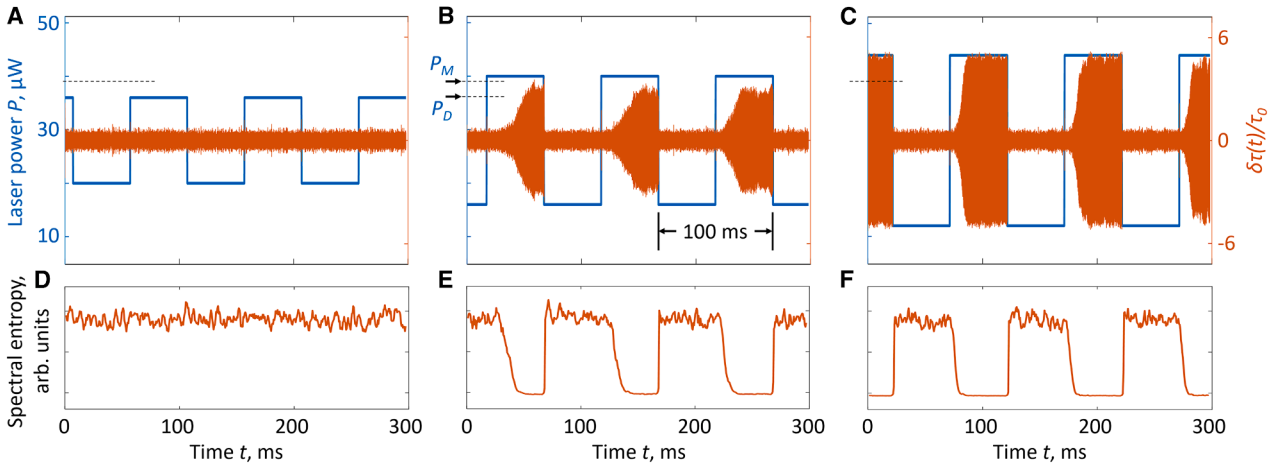

Figure 2. Dynamics of optically induced spontaneous mobilization and demobilization transitions in the metamaterial continuous time crystal

(A–C) Relative transmissivity change $\delta\tau(t)/\tau_0$ of the metamaterial array (orange lines, right-hand axis) related to the out-of-plane motion of nanowires as a function of time, while incident laser power is switched between higher and lower levels (blue lines, left-hand axis).

(D–F) Corresponding dynamics of transmissivity spectral entropy.

reduces to a single fixed cycle, and an ergodicity collapse is observed.[26,27]

Importantly, the results presented in Figure 3 also demonstrate that continuous time-translation symmetry is broken by the mobilization transition through the randomness of the phase of the mobilized state. Indeed, Figure 3D illustrates that the system "winds up" to the mobilized state through a different phase trajectory in each cycle with an arbitrary initial phase prescribed by thermal fluctuations, while the distribution of points in Figure 3E illustrates the random phase of oscillation at a fixed interval after the incident laser intensity exceeds the mobilization threshold.

Modeling the transient dynamics of mobilization and demobilization

The main features of the metamaterial time crystal's mobilization and demobilization dynamics can be understood from a model of nonreciprocally coupled linear oscillators. We consider a set of 26 oscillators, with thermal motion at non-zero temperature

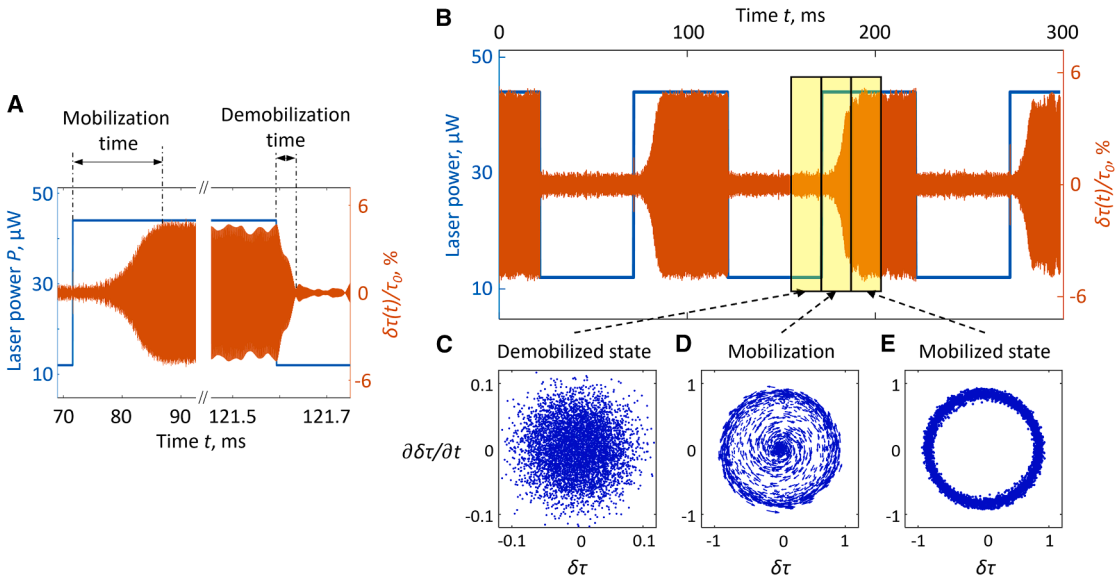

Figure 3. Transient mobilization and demobilization times

(A and B) Transmissivity oscillation dynamics during mobilization and demobilization.

(C–E) Transmissivity phase diagrams (C) of the disordered state, (D) during mobilization, and (E) of the mobilized time-crystal state. Phase diagrams are derived from analysis of the segments indicated by the yellow shaded boxes in (B) over hundreds of repeated mobilization cycles.

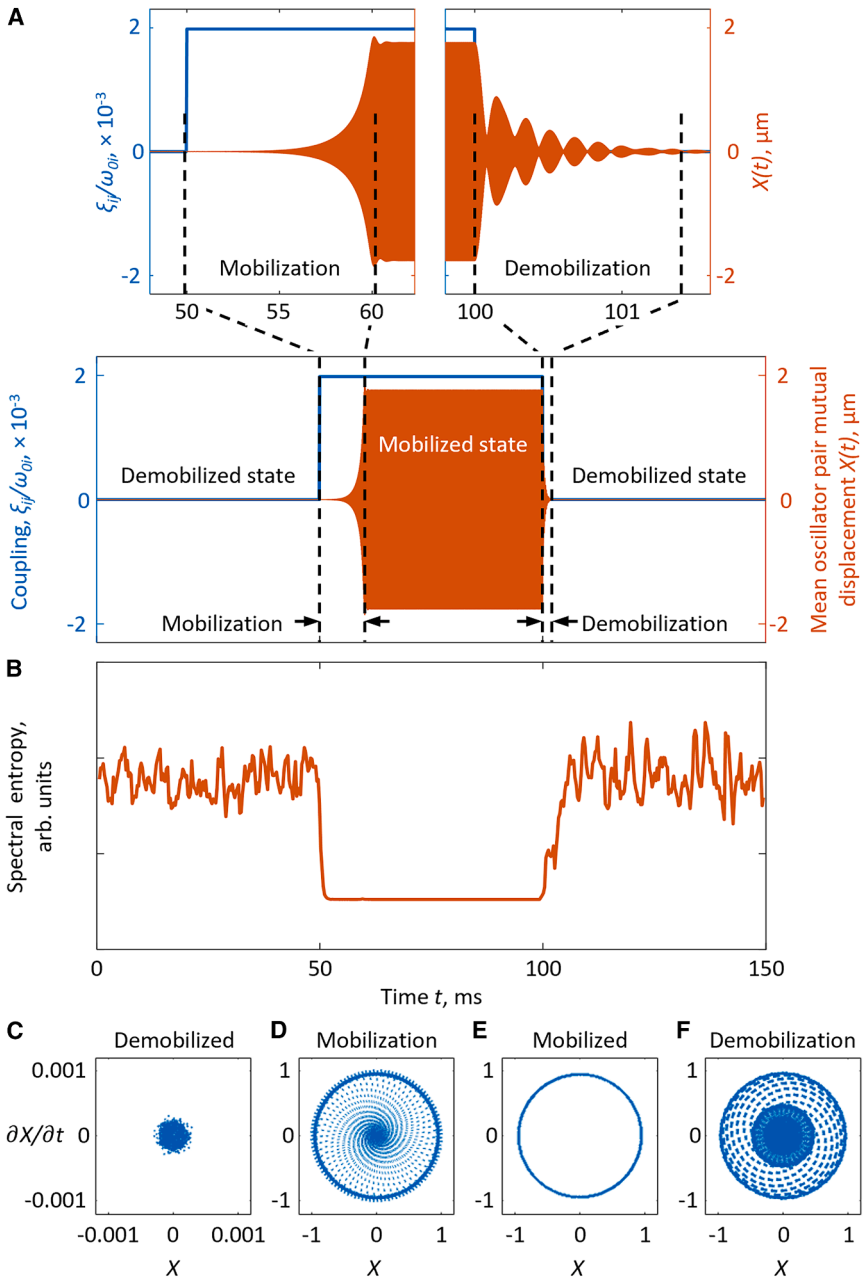

Figure 4. Dynamics of an ensemble of 26 thermally driven, nonreciprocally coupled Langevin oscillators

(A and B) Time dependence of (A) the mean mutual displacement between oscillator pairs *X(t)* (orange traces) and (B) displacement spectral entropy as coupling strength (blue traces in A) is cycled through step changes from zero to a value above the array's synchronization threshold and back.

(C–F) Displacement phase diagrams (C) for the demobilized state, (D) during mobilization, (E) for the mobilized state, and (F) during demobilization.

accounted for by assuming that they are connected to a common bath (in the experiment, the frame of the nanowire array) at temperature *T*. The dynamics of such oscillators near their equilibrium positions are described by the Langevin model:

$$\ddot{x}_i + \gamma_i \dot{x}_i + \omega_{0i}^2 x_i + \sum \xi_{ij}(x_i - x_j) + \alpha x^2 + \beta x^3 = \sqrt{\frac{2k_B T \gamma_i}{m_i}}\,\eta_i(t), \quad \text{(Equation 1)}$$

where x is the position; ω_{0i}, m_i, and $\gamma_i = \frac{\omega_{0i}}{Q_i}$ are the natural frequencies, masses, and damping parameters of the oscillators; and $\eta_i(t)$ are normalized white-noise terms. Light-induced coupling between the oscillators is described by the parameter ξ_{ij}. Here, we assume the nonreciprocal nature of coupling $\xi_{ij}/\omega_{0i} = -\xi_{ji}/\omega_{0j}$ and take into account next-neighbor coupling only; thus, $\xi_{ij} = \xi\delta_{i,j\pm1}$, where δ_{ij} is the Kronecker delta.

The synchronization process relying on nonreciprocal forces starts from incoherent thermal oscillations with a characteristic initial amplitude of ~300 pm and does not require any intrinsic nonlinearity in the system. However, the nonlinearity of the restoring force on the oscillators becomes noticeable when the amplitude becomes comparable to the nanowire thickness (~50 nm) and it constrains the exponential growth of oscillation amplitude. This is known as the "geometric nonlinearity" of a doubly clamped, bilayer bar[28] and is accounted for by nonlinear terms in the restoring force $\alpha x^2 + \beta x^3$ (where α and β are constants).

We analyze the behavior of this system by numerically solving Equation 1 using oscillator parameters close to those of the experimental system: masses m_{2i-1} = 1.4 pg and m_{2i} = 0.78 pg; natural frequencies ω_{0i} are taken to be randomly distributed alternately around $2\pi \times 873$ and 881 kHz (either side of an ensemble mean frequency $\omega_c = 2\pi \times 877$ kHz, to satisfy conditions for the establishment of a predator-prey dynamic[15]); and $\gamma_i = \omega_{0i}/Q$, where Q = 1,000. We assume $\alpha = 3.2 \times 10^4\ \omega_c^2$ and $\beta = 10^9\ \omega_c^2$, and impose step changes in the value of the coupling coefficient ξ_{ij} to mimic the action of turning incident laser light (cf. optical interaction between nanowires) on and off.

The results of this modeling are shown in Figure 4, in terms of the mean mutual displacement between oscillator pairs $X(t) = \frac{1}{13}\sum_{i=1}^{i=13}(x_{2i}(t) - x_{2i-1}(t))$ as a surrogate for the experimentally measured time-dependent normalized transmissivity $\delta\tau(t)/\tau_0$ (Figure 3), and spectral entropy. The results correlate exceptionally well with experimental data,

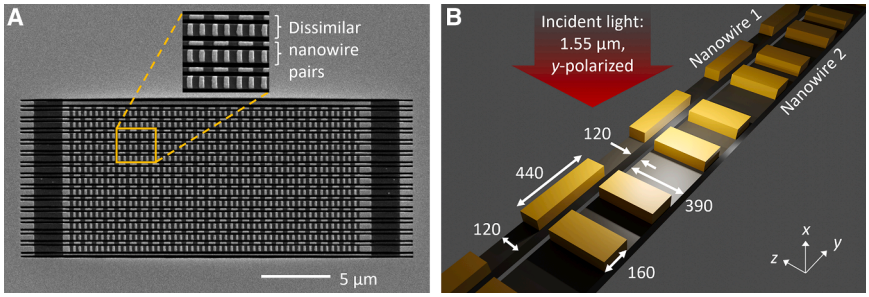

Figure 5. Nanowire metamaterial design

(A) Scanning electron microscope image of a nano-opto-mechanical (time-crystal) metamaterial.

(B) Artistic schematic of one pair of dissimilar silicon nitride nanowires decorated with plasmonic nanorods (dimensions in nm).

qualitatively reproducing all the key features of the mobilization and demobilization transitions. Considered together, they demonstrate that nonreciprocal coupling is the main mechanism of mobilization in the metamaterial time crystal.

Several light-induced mechanisms contribute to the nonreciprocal, mutual position-dependent interaction between nanowire oscillators, emerging from the radiation pressure and absorption of light scattered by dissimilar parts of the plasmonic metamolecules located on neighboring nanowires: Radiation pressure acts directly on the oscillators,[15] while the dissipation of light leads to anisotropic expansion of the bilayer structure through thermal and optical striction,[29] resulting in nonreciprocal out-of-plane forces. These nonreciprocal interactions depend on the wavelength of light and are resonantly enhanced in plasmonic metamolecules.

DISCUSSION

We present the first experimental study of the dynamics of mobilization and demobilization transitions between the states of incoherent thermal motion and persistent synchronized oscillation in a nano-opto-mechanical metamaterial time crystal—an array of alternately dissimilar nanowires with variable, optically controlled nonreciprocal coupling between them.

By analyzing the time-domain response of the system, we have shown experimentally that the speed of mobilization, from incoherent thermomechanical fluctuation to synchronized oscillation of the array, increases with incident light intensity (Figures 2B and 2C) and that the mobilization transition is slower than the intensity-independent demobilization transition to the disordered state, which occurs upon withdrawal of illumination and which is controlled by damping and dephasing of the inhomogeneous broadened nanowire oscillator ensemble. By resolving the temporal evolution of the phase portrait of the system, we have shown that the mobilization transition is accompanied by ergodicity breaking, whereby, with increased optical coupling, the oscillation frequencies of individual nanowires converge to a single-phase state.

We note that this mechanism of realizing a continuous time crystal requires the presence of two dissimilar components—in experiment, nanowires supporting different parts of the plasmonic metamolecules—exhibiting predator-prey dynamics that are characteristic of a wide variety of systems in which nonreciprocity-driven symmetry breaking, pattern formation, hysteresis, and phase transitions are observed.[2–4,6,7] Interestingly, the observed decrease in the entropy of the optically mobilized time-crystal state may be compared with the entropy dynamics of plants growing under the influence of the sun, wherein disordered matter evolves into highly organized living matter,[30] illustrating the recently proposed view[3] that nonreciprocal interactions may have played a critical role in the matter-to-life transition. Indeed, in manifesting such characteristics, optically controlled continuous time crystals driven by nonreciprocal interaction forces may represent a useful platform for modeling the matter-to-life transition.

Time crystals on nano-opto-mechanical platforms will be of use in the study of the classical dynamics of many-body systems in the strongly correlated regime, complementing cold atom and spin quantum platforms. Structured illumination of the interacting oscillators of a time crystal may also present an interesting opportunity for implementing all-optical neural networks and reservoir computer architectures. Finally, we would argue that nano-opto-mechanical continuous time crystals represent a material platform suitable for developing various components of optical timetronic functionality,[24] derived from their ability to spontaneously break time-translation symmetry and ergodicity.

METHODS

Nanowire metamaterial fabrication and experimental characterization

The array of nanoscale oscillators was manufactured by focused ion beam milling from a 50-nm-thick, low-stress (<250 MPa) silicon nitride membrane (Norcada) coated with a 50 nm layer of thermally evaporated gold. It comprises twenty-six 28-μm-long, alternately 120- and 390-nm-wide nanowires (i.e., thirteen pairs of dissimilar nanowires), supporting Π-shaped plasmonic metamolecules, each consisting of three gold nanorods—one on the narrow nanowire with its long axis parallel to that of the wire and two in perpendicular orientation on the wider wire (Figures 5A and 5B).

For optical characterization, i.e., of time-crystal transition dynamics, the sample was mounted in a vacuum cell at $\sim 10^{-4}$ mbar to exclude viscous (air) damping of nanowire movement. The array was illuminated with a single beam of laser light at a wavelength of 1.55 μm (close to the plasmonic absorption resonance of the metamolecules), used both to monitor the transmissivity of the array and to induce transitions to the synchronized oscillatory (mobilized, time-crystal) state. Light was normally incident on the array, polarized parallel to the nanowires, and

focused to a spot with a full-width half-maximum diameter of ~5 μm. Incident power was controlled by an electro-optic modulator. Transmitted light intensity was monitored using an InGaAs photodiode with 125 MHz bandwidth.

RESOURCE AVAILABILITY

Lead contact

Requests for further information should be directed to and will be fulfilled by the lead contact, Kevin F. MacDonald (kfm@orc.soton.ac.uk).

Materials availability

The data from this paper can be obtained from the University of Southampton ePrints research repository: https://doi.org/10.5258/SOTON/D3489.

Data and code availability

Any additional information required to reanalyze the data reported in this paper is available from the lead contact upon request.

ACKNOWLEDGMENTS

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AUTHOR CONTRIBUTIONS

The project was conceived by N.I.Z. Experimental work was undertaken by T. L. and analytical and computational modeling by V.R. and J.L. All authors contributed to the analysis and interpretation of results and writing of the manuscript. Work was supervised by K.F.M. and N.I.Z.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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