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Photonic metamaterial analogue of a continuous time crystal

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Time crystals are an eagerly sought phase of matter with broken time-translation symmetry. Quantum time crystals with discretely broken time-translation symmetry have been demonstrated in trapped ions, atoms and spins whereas continuously broken time-translation symmetry has been observed in an atomic condensate inside an optical cavity. Here we report that a classical metamaterial nanostructure, a two-dimensional array of plasmonic metamolecules supported on flexible nanowires, can be driven to a state possessing all of the key features of a continuous time crystal: continuous coherent illumination by light resonant with the metamolecules' plasmonic mode triggers a spontaneous phase transition to a superradiant-like state of transmissivity oscillations, resulting from many-body interactions among the metamolecules, characterized by long-range order in space and time. The phenomenon is of interest to the study of dynamic classical many-body states in the strongly correlated regime and applications in all-optical modulation, frequency conversion and timing.

Isotropic homogeneous matter is invariant under space-translation symmetry while in crystals with periodic atomic lattices that symmetry is broken. In nature, these symmetry-breaking crystalline states with long-range order are achieved spontaneously through a phase transition (for example, from water to ice). In recent years, the physics community has been captivated by the newly described phase of matter known as a 'time crystal', with broken time-translation symmetry, analogous to conventional crystals in which space-translation symmetry is broken. A time crystal, as originally proposed by Wilczek[1], is a quantum many-body system whose lowest-energy state is one in which the particles are in continuous oscillatory motion. Although it has been shown that such closed systems, breaking continuous time-translation symmetry by exhibiting oscillatory dynamics, are prohibited by nature[2], discrete time crystals that show time-translation symmetry breaking imposed by an external modulated parametric drive have been realized on various platforms, including an interacting spin chain of trapped atomic ions[3]; a disordered ensemble of spin impurities in diamond[4]; a crystal lattice of nuclear spins in an ammonium dihydrogen phosphate crystal[5]; an ensemble of ultracold atoms, in which the periodic structure in both space and time is observed[6]; a Bose–Einstein condensate of magnons, in which an oscillating magnetic field induces coherent spin precession at a frequency incommensurate with the drive frequency[7]; a Kerr non-linear optical microcavity pumped by two lasers, with self-injection locking[8] and a curved array of interacting microwave waveguides[9]. Key signatures of the many-body-localized discrete time crystal state have also been observed on a quantum simulation platform based on individually controllable carbon-13 nuclear spins in diamond[10], and on arrays of superconducting qubits in a quantum processor[11,12], whereas a prethermal discrete time crystal (a non-equilibrium driven phase without disorder) has been realized in a trapped-ion quantum simulator[13].

Recently, a quantum time crystal that breaks time-translation symmetry continuously has been observed in an atomic Bose–Einstein condensate inside an optical cavity, manifested in the emergence of spontaneous oscillations of the intracavity photon number under optical pumping[14]. In this experiment, the pump laser was operated continuously, thereby respecting continuous time-translation symmetry while inducing a transition to the symmetry-broken state.

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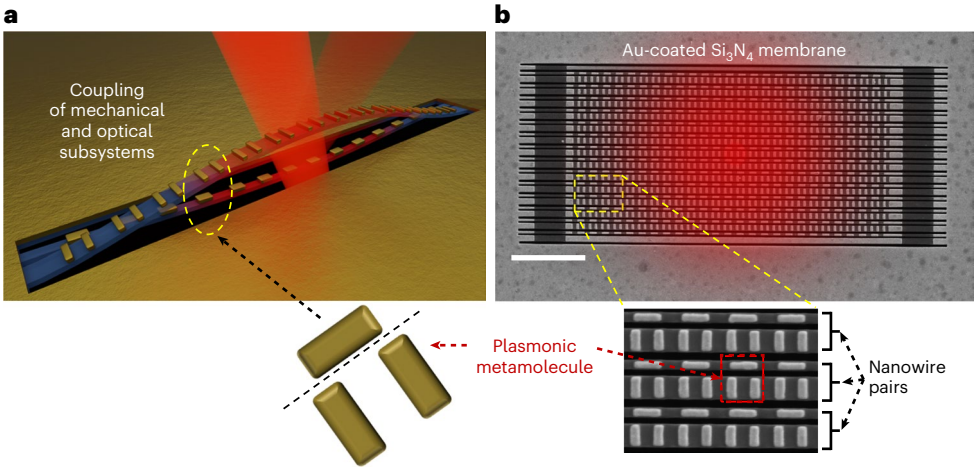

Fig. 1 | Photonic metamaterial analogue of a continuous time crystal. A two-dimensional array of plasmonic metamolecules is supported by nanowires cut from a semiconductor membrane. **a**, Artistic impression of the basic building block of the crystal—a pair of silicon nitride nanowires decorated with plasmonic (gold) metamolecules. **b**, Scanning electron microscope image of the entire two-dimensional array. Illumination with coherent light (as indicated by the schematically overlaid laser spot) induces a transition to a state of persistent synchronized nanowire oscillations. Scale bar, 5 µm.

This atomic system realizes the spirit of Wilczek's original proposal more closely than discrete time crystals and could be a platform for exploring entanglement and topological phases in arrays of levitated particles and quantum simulations.

A defining characteristic of a continuous time crystal is a spontaneous transition, in a many-body system, to a robust oscillatory state in reaction to a time-independent external stimulus. Here we show that a two-dimensional periodic lattice of plasmonic metamolecules supported on doubly clamped nanowire beams cut from a semiconductor membrane (Fig. 1) spontaneously transitions, at room temperature, to a state characterized by persistent optical transmissivity oscillations when illuminated by coherent light that stimulates interaction among the metamolecules. Above a threshold level of incident optical power, the spectrally dispersed thermal fluctuations of the individual nanowires become spatially coherent synchronous oscillations over the illuminated ensemble.

Here, it should be noted that the term 'photonic time crystal' bears some ambiguity. In the condensed-matter physics and cold atom communities, and in the present work, it denotes a stable phase of matter that spontaneously breaks continuous time-translation symmetry through multi-body interactions. However, in the photonics community the terminology is often used more broadly in reference to wave processes in active systems relying on externally induced modulation of their constitutive parameters (reviewed in ref. 15). For instance, wave amplification inside the momentum bandgap of a modulated microwave metamaterial has recently been demonstrated[16], while temporal reflections and broadband frequency translations have been demonstrated in a switched transmission-line metamaterial[17].

Results

The experimental sample was fabricated by focused ion beam milling from a gold-coated silicon nitride nano-membrane (Fig. 1b). The plasmonic metamolecules—Π-shaped arrangements of three gold nanorods—are supported on pairs of neighbouring nanowires in such a way that the nanowires' mutual displacement (Fig. 1a) strongly affects the metamolecules' resonant plasmonic scattering properties. This coupling between mechanical and optical properties of the lattice provides opportunity for detecting the state of the lattice optically by monitoring its transmissivity. Indeed, it was shown recently that even picometric thermomechanical fluctuations in such structures can measurably modulate scattered and transmitted light[18].

In such an array, the nanowires can be described by set of N coupled classical oscillators with indices i and j each governed by the following equations of thermally driven motion:

$$\ddot{x}_i + \Gamma_i \dot{x}_i + \omega_{0i}^2 x_i + \alpha x_i^2 + \beta x_i^3 + \sum_{i,j=1}^{N} \xi_{ij}(I)(x_i - x_j) = \sqrt{2k_B T \Gamma_i / m_i}\,\eta_i(t) \quad (1)$$

According to the Langevin treatment of thermally driven oscillators, the right-hand side of the equation is the time-dependent thermal force experienced by the oscillator, which depends on the dissipation factor Γ_i through the fluctuation-dissipation theorem. $\eta_i(t)$ is a normalized white noise term, m_i is the effective mass of the nanowire, k_B is the Boltzmann constant, T is the temperature and ω_{0i} is the nanowire's natural angular frequency of oscillation. A nanowire clamped at both ends exhibits 'geometric non-linearity' (terms αx_i^2 and βx_i^3) that becomes notable when the amplitude of oscillation approaches the thickness of the nanowire[19]. Owing to fabrication imperfections and intrinsic variations of stress in the membrane in the present case, the fundamental frequencies of the nanowires ω_{0i} are dispersed within a range of about 2% around a central frequency $\omega_0 = 2\pi \times 0.99$ MHz.

It has been shown that in absence of noise, a pair of interacting non-linear oscillators can be driven into the discrete time crystal state by parametric modulation of their natural frequencies[20]. Here we experimentally demonstrate a different route that leads to the continuous time crystal state: the combination of noise and interaction among oscillators induced by continuous illumination triggers a first-order dynamic phase transition to a state in which time-translation symmetry is broken. When the lattice of plasmonic metamolecules is illuminated, an optically controllable interaction between nanowires, described by the term $\xi_{ij}(I)(x_i - x_j)$ in the above equation of motion, emerges through dipole–dipole coupling of the induced plasmonic (gold nanorod) excitations. Here the coupling coefficient is a function of light intensity I and the mutual position of the oscillators.

The array is illuminated with a single continuous beam of laser light at a wavelength of 1,550 nm (close to the plasmonic absorption resonance of the metamolecules; Supplementary Information), which is used both to monitor the transmissivity of the array and to induce the synchronized oscillatory state. The sample is housed in a low vacuum optical cell with pressure maintained at around 10^{-4} Torr to suppress damping of nanowire movements. Light is normally incident on the array, polarized parallel to the nanowires and focused to a spot with a

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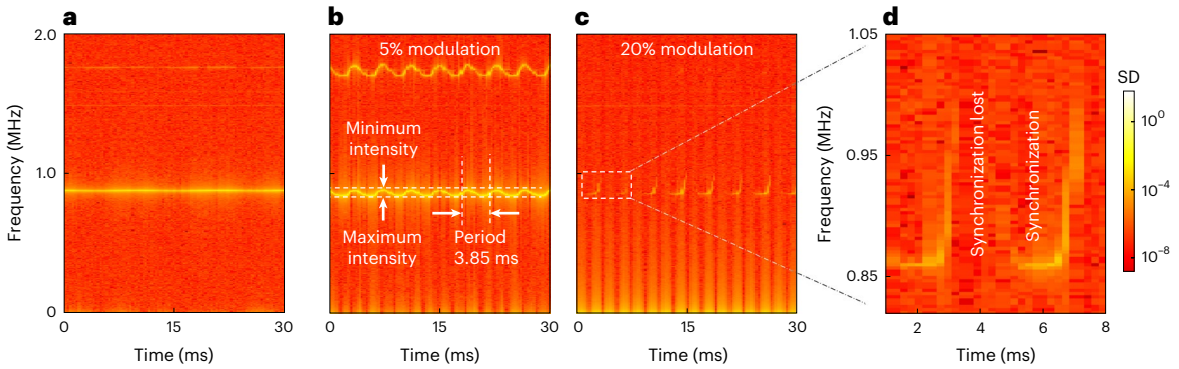

Fig. 2 | Persistence, stability and interruption of synchronized oscillations. **a**–**d**, Evolution of the spectrum of transmissivity oscillations (SD, spectral density): with time-invariant incident optical power above the threshold of synchronization (**a**), whereby synchronized oscillation persists indefinitely and the frequency of oscillation does not change; with incident power sinusoidally modulated to a depth of 5% (from the $P = 124.3$ μW level, at 260 Hz) (**b**), whereby synchronized oscillation is maintained at a frequency that adiabatically depends on incident power; at a higher level of incident optical power modulation (20%) (**c**), whereby synchronization is periodically suppressed and recovered, giving rise to transmissivity oscillation bursts, shown in closer detail in **d**.

full-width half-maximum diameter of roughly 5 μm. The transmitted light intensity is monitored by an InGaAs photodetector.

At low laser power (≤tens of μW), the transmissivity spectrum contains several overlapping peaks of small amplitude at frequencies just below 1 MHz corresponding to oscillations of the individual illuminated nanowires. This is typical of an ensemble of nanowire oscillators with uncorrelated thermomechanical dynamics of characteristic amplitude $x = \frac{1}{\omega_{0i}}\sqrt{\frac{k_B T}{m_i}}$, roughly 250 pm. While adiabatically increasing laser power, we initially observe red shifting of the spectral peaks, due to thermal expansion of the nanowires induced by laser illumination, as described in ref. 21. At around 130 μW, the onset of synchronization is observed in a narrowing of the spectrum. Further increasing the laser power leads to spontaneous synchronization of the nanowires' oscillation, manifested in the emergence of a single narrow peak in the transmissivity modulation spectrum, with an amplitude four orders of magnitude larger than the peaks associated with the individual nanowires' uncorrelated thermomechanical fluctuations. While adiabatically decreasing laser power, we observe hysteretic loss of synchronization and reversion of the transmissivity modulation spectrum to its low-power form (that is, comprising several small amplitude peaks) below about 115 μW, revealing the first-order nature of the synchronization phase transition.

One of the main features of a continuous time crystal is that, once initiated by an external stimulus, oscillations persist over arbitrarily long times and this is true of our experimental system; Fig. 2a shows that the spectrum of synchronized transmissivity oscillations is invariant as a function of time for a constant level of incident optical power (ramped initially from zero to $P = 150$ μW, whereby a strongly synchronized state (SSS) is achieved and then held at $P = 124.3$ μW). The observed transmissivity oscillations are also stable against small perturbations; Fig. 2b shows slow (adiabatic) modulation of the transmissivity oscillation frequency driven by thermal expansion and contraction of the nanowires with modulation of incident power (and thereby nanowire temperature). Here, the incident light intensity is modulated by 5% at a frequency of 260 Hz, and the temperature of the nanowires adiabatically increases and decreases with laser power as their cooling time of roughly 85 μs is much shorter than the 3.85 ms period of intensity modulation. Here, we can also see that the periodic modulation of transmissivity is anharmonic: the transmitted light has a spectral component of modulation at the second harmonic of the main out-of-plane mode of the structure. This is explained by the geometric non-linearity of nanowire motion: for nanowires of asymmetric cross-section (semiconductor beams decorated on one side with plasmonic nanorods), both quadratic and cubic terms are present in the equation of motion (terms αx_i^2 and βx_i^3 in equation (1)).

When the incident optical power is more strongly modulated (20%), periodic suppression and recovery (bursts) of transmissivity modulation are observed, as illustrated in Fig. 2c,d. In both cases (5 and 20% incident power modulation), periodic shifting of the nanowires' natural frequencies in response to modulation of the incident power leads to their synchronous oscillations being periodically chirped. As such, there is a low-frequency spectral component extending from zero to the chirp frequency, which is visible as a pattern of low-frequency 'fringes' on the logarithmic scale of Fig. 2b,c. The fringes increase in brightness with increasing rate of change of laser power, and disappear when the power reaches maxima and minima; hence, two fringes are observed for each period incident laser power modulation.

To illustrate that synchronized oscillations have arbitrary phase each time a burst is generated, as is characteristic of a continuous time crystal[14], we analysed their phase distribution. Fig. 3a shows a typical time domain trace of such a burst. We find that the phase of spontaneous oscillation, relative to the cycle of incident light modulation, is indeed randomly distributed over the $(0, 2\pi)$ range, as illustrated in Fig. 3b where the real and imaginary parts of the amplitude at 855 kHz—the frequency at which an SSS is achieved (Fig. 4c)—are presented for 155 consecutive bursts. These data indicate that the phase of nanowire oscillations relative to that modulation takes random values.

Figure 4 presents analysis of the structure of a single spontaneous burst of oscillation resulting from a cycle of 20% decrease and recovery in incident laser power (as presented in Fig. 2d). To evaluate how the peak frequency and amplitude of the burst evolves in time and with incident laser power, we evaluated Fourier spectra of transmissivity for 25 consecutive time intervals over one incident laser power modulation period. This analysis yields values of the peak amplitude, width and frequency of oscillation spectral density (Fig. 4a–c) at the average value of incident laser power for each interval.

We observe that in the dynamic regime of cyclically increasing and decreasing incident laser power, the spontaneous transition to and from synchronization has all of the characteristics of a first-order phase transition, where the oscillation spectral density peak amplitude can be used as an order parameter for synchronization (here and below, terminology for states of the oscillator ensemble is adopted from ref. 22). With increasing laser power (orange symbols in Fig. 4) the array of oscillators is initially in an incoherent state (IS), characterized by small amplitude oscillations, wherein measurements of peak frequency and width are not possible. When the incident laser power exceeds roughly 130 μW, a weakly synchronized state (WSS) emerges and the amplitude

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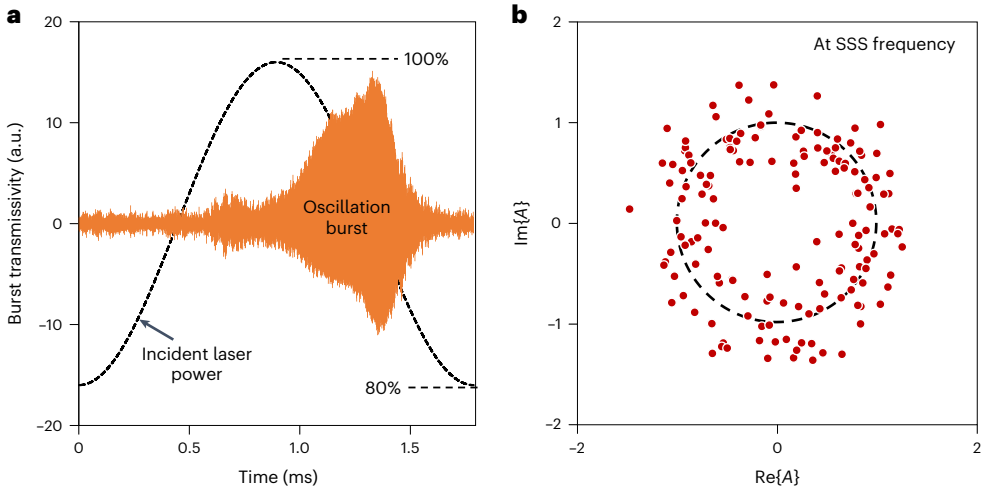

Fig. 3 | Synchronization phase. a, Time domain trace of a single transmissivity oscillation burst (orange line) overlayed on a trace showing the time dependence of incident laser power (in the 20% modulation regime, dashed black line). a.u., arbitrary units. **b**, Imaginary versus real part of spectral amplitude (A) at 855 kHz, the frequency at which an SSS is achieved, from the Fourier spectra of 155 consecutive bursts of transmissivity oscillation over intervals equal to the period of incident light intensity modulation, as illustrated in Fig. 3a. Incident light intensity is modulated at 560 Hz.

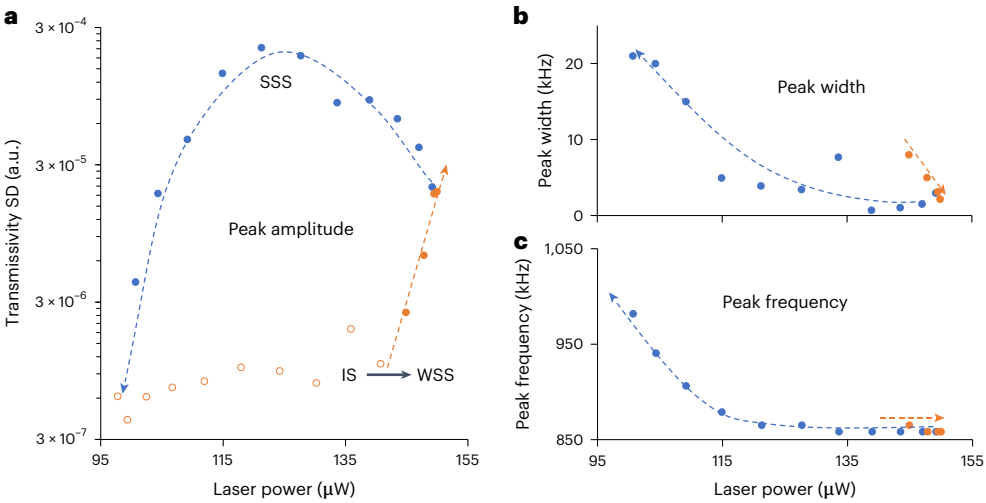

Fig. 4 | Synchronization as a first-order phase transition process. a–**c**, Analysis of a burst of metamaterial array transmissivity oscillation during a single cycle of 20% incident laser power modulation: the peak amplitude (**a**), spectral width (**b**) and central frequency (**c**) of transmissivity spectral density (SD) for increasing (orange symbols) and decreasing (blue) incident laser power. Oscillatory regimes referred to as the IS, WSS and SSS are annotated in **a**. For increasing laser powers from zero up to the IS-to-WSS transition, the small level of transmissivity modulation does not allow reliable measurements of oscillation frequency or peak width. As such, no symbols are plotted in **b** and **c** in this range, and hollow symbols are plotted in **a**.

of oscillation spectral density grows exponentially, as indicated by the orange dashed line in Fig. 4a. At an incident laser power of roughly 150 μW, an SSS is achieved, which is characterized by the oscillation spectrum collapsing to a line of <2 kHz width. With decreasing power (blue symbols in Fig. 4a), the SSS state persists until the incident power falls below around 100 μW and the system returns to the IS, completing a full hysteresis loop as is characteristic of a first-order phase transition. A well-defined stable frequency of oscillation roughly 855 kHz that is achieved in the SSS regime, remains invariant and resilient to the variations in the level of light-induced heating until power drops to roughly 120 μW (Fig. 4a,c), and is stable from burst to burst with variance of 1.1 kHz. The pronounced dependence of the frequency of transmissivity oscillation on incident power below 120 μW (Fig. 4c) is due to thermal contraction of the nanowires induced by decreasing laser illumination[18].

Discussion

We argue that the underlying dynamics of the observed light-assisted phase transition to a persistent, high amplitude transmissivity oscillation regime, the main feature of a time crystal, can be interpreted as resulting from the synchronization of the ensemble of noisy thermomechanical nanowire oscillators via light-induced coupling $\xi_{ij}(I)$. Indeed, as introduced by Kuramoto[23], populations of interacting self-oscillators with distributed natural frequencies can show remarkable transitions from a disordered state to an ordered one as the strength of interactions is varied and exceeds a certain threshold: a macroscopic number of elements begin to be mutually entrained with a common frequency. Within this general approach, Tanaka et al. have shown[22] that the synchronization of a periodically driven system of oscillators with inertia exhibits a first-order phase transition to synchronization from an IS to a hysteresis loop between two macroscopic states, a WSS and a strongly

coherent synchronized state (SSS). Later, Hong et al. analysed this phenomenon with particular attention to the effects of noise[24]. They found that the coupling needed to achieve a phase transition increases, and the hysteresis collapses, when thermal noise coming into the system increases. Indeed, simple energy considerations show (Supplementary Information) that the laser power threshold for synchronization should increase with temperature. A regime of synchronization in an ensemble of globally coupled phase oscillators with distributed intrinsic oscillator frequencies and external independent noise forces—a system closer to the present experiment—has also been investigated, indicating a non-equilibrium transition between desynchronized and synchronized states and the existence of bistability[25,26].

The components of the metamolecules—plasmonic particles located on different nanowires—behave as induced dipoles driven by the optical field. If the particles are identical (as on alternate nanowires) and the induced dipoles are in phase, the interaction between them is reciprocal and $\xi_{ij}(I) = \xi_{ji}(I)$. If the particles are different in size and/or geometry (as on neighbouring nanowires), so are their resonant properties and induced dipoles. In this case, as the light-induced interaction between particles may include a combination of a conservative gradient force and a non-conservative radiation pressure force, the interaction between two different particles can be non-reciprocal: $\xi_{ij}(I) = -\xi_{ji}(I)$. The *actio et reactio* equality is broken, leading to the appearance of a non-reciprocal interaction force in the equation of motion (1). The non-conservative contribution emerges from the radiation pressure induced by the scattered fields, which constantly pumps energy into the system, and thus cannot be derived from a Hamiltonian[27]. Following from earlier theoretical works by Sukhov et al.[28] and Karasek[29], such non-reciprocal light-induced interaction has recently been observed experimentally with optically trapped particles[27]. From the theory of coupled oscillators, it is known that non-reciprocal coupling does not split resonant frequencies but does lead to an increase in oscillation amplitude. We therefore argue that non-reciprocal coupling may play a role in the synchronization of ensembles of oscillators.

Our ensemble of nanowire oscillators exhibiting thermal motion and interacting with help of an external light field can be seen as a classical analogue of an ensemble of quantum emitters interacting with a common light field and exhibiting Dicke superradiance[30]. If the wavelength of the light is much greater than the separation of the emitters, then the emitters interact with the light collectively and coherently emit light with intensity proportional to N^2 (superradiance), very differently from the emission with intensity proportional to N observed for a group of independent emitters. Indeed, the relationship between the Dicke superradiant state and discrete time crystal ordering was recently theoretically generalized in terms of the Landau theory of phase transitions[31] and was also noted in regard to the recently demonstrated quantum continuous time crystal[14].

In summary, we have demonstrated an artificial photonic material—a two-dimensional array of plasmonic metamolecules supported on flexible nanowires—that can be driven to a state showing all the defining characteristics of a continuous time crystal—a new state of matter that continuously breaks time-translation symmetry: (1) time-independent excitation, in the form of illumination with coherent light that is resonant with the plasmonic modes of the metamolecules, spontaneously triggers strong periodic oscillation of optical transmissivity; (2) the oscillations result from many-body interactions among metamolecules coupled by electromagnetic dipole–dipole interactions (induced by the incident light), whereby the individual thermal oscillations of nanowires are replaced by coherent, superradiant-like motion of the illuminated ensemble; (3) the transition to the coherent oscillatory regime has the nature of a first-order phase transition; (4) the transmissivity oscillations are robust to small perturbations and the phase of the oscillation is random for different realizations and (5) the state shows long-range order in space, as manifested in synchronization of the illuminated ensemble, and long-range order in time, as seen in the robust indefinite persistence of synchronous oscillation.

Apart from its intrinsic conceptual interest, our results and the simplicity and control achieved with our nano-opto-mechanical platform, offer a new class of dynamical many-body system that expands the concepts of long-range order and spontaneous symmetry breaking into the time domain. It paves the way towards comprehensive studies of dynamic many-body states in the strongly correlated regime in a classical system that complements the cold atom and spin platforms where many-body quantum states of bosonic or fermionic matter can be investigated. These studies can be facilitated by direct access to the nanowire positions using recently developed atomic scale techniques for detecting localization and movement of nanoscale strings and cantilevers[32,33]. The dynamics of transitions from the stochastic and ballistic thermal motion of individual nanowires into the collective superradiant state at difference time scales and at various ambient temperatures are also of interest for nanoscale thermodynamics[32].

Micro- and nanomechanical metamaterials have been demonstrated across the electromagnetic spectrum from microwave to terahertz and optical frequencies using a variety of materials, from silicon to high-index dielectrics and plasmonic metals, fabricated using focused ion beam milling, optical and electron beam lithography. The design of metamolecules determines the quality factors of resonances and bandwidth of such devices and as such the platform can be readily adapted to different applications. Step-like transitions to a robust superradiant mode induced by light may be used in optically controlled modulators and frequency mixers not yet known among photonic technologies. Such triggering could also be employed in new all-optical timing applications where periods of oscillation are counted after triggering, and the random phase of spontaneous oscillations may be exploited in all-optical pseudo-random number generators based on a classical platform, in contrast to generators exploiting the intrinsic randomness of quantum mechanics.

Online content

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-023-02023-5.

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Data availability

For the purpose of open access, the authors have applied a creative commons attribution (CC BY) licence to any author accepted manuscript version arising. The data from this paper can be obtained from the University of Southampton ePrints research repository: https://doi.org/10.5258/SOTON/D2540. Source data are provided with this paper.

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Author contributions

The project was conceived by N.I.Z. Experimental work was undertaken by T.L. All authors contributed to analysis and interpretation of data and writing of the paper. Work was supervised by J.Y.O, K.F.M. and N.I.Z.

Competing interests

The authors declare no competing interests.

Additional information

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