SOFT ACTUATORS

Synchronized dancing under light

Coupled liquid-crystalline network oscillators incorporating light-responsive molecules show synchronized motion when stimulated by light. This behaviour resembles that of synchronized clock pendulums and may find uses in advanced soft robotics applications.

Yanlei Yu

Synchronized oscillations, or coupled oscillations, were first reported in 1665 by the Dutch physicist Christiaan Huygens, the inventor of the pendulum clock[1]. Huygens noted that two identical pendulum clocks mounted on the same wooden bar often swung at the same frequency but in opposite directions. This interesting synchronization phenomenon named in his honour results from the energy transmission of the two clocks through the connected rigid wooden structure[2]. Inspired by this process, oscillators and related robotic systems have been developed, most of which are based on energy propagation through rigid materials. Now, writing in *Nature Materials*, Vantomme and colleagues demonstrate both experimentally and theoretically that such energy transmission also occurs in fully soft materials, namely, in a fork-like film made from light-responsive liquid-crystalline networks (LCNs) that displays collective synchronized oscillations upon light irradiation[3].

Azobenzene-containing LCNs are common photoresponsive soft materials that have potential in advanced devices and systems such as soft robots and mixed reality devices[4,5]. The ordered alignment of the liquid-crystal mesogens in the LCNs promotes the photo-deformability with predetermined topology[6,7]. For example, LCN actuator films, whose molecular alignment gradually changes from perpendicular to the long axis at the bottom side to parallel at the upper side, are able to continuously oscillate upon light irradiation owing to the shadowing effect[8]. The unique molecular alignment on the surface allows the actuators to bend away when exposed to light, while the newly light-exposed part will bend the actuators in the opposite direction. With the aid of these LCN actuators, Vantomme and co-workers have developed a general approach to investigate the energy transmission between two LCN actuators during photo-actuated oscillations.

In their study, two strip-like actuators connected by the same material were fabricated by cutting a rectangular piece of an LCN film in the middle (Fig. 1a). Upon light irradiation, the two actuators produce collective, synchronized in-plane or anti-plane oscillations with the same period and amplitude (Fig. 1b,c). In particular, owing to the presence of the coupling joint, energy transmission occurs between the two LCN actuators, as described for rigid systems such as the synchronization of pendulum clocks reported by Huygens. The oscillation frequency of the coupled soft actuators in anti-phase synchronization (~9.5 Hz) is slightly higher than the oscillation frequency in in-plane synchronization (~8.5 Hz). On the contrary, two independent LCN actuators in close proximity to each other show individual oscillations with no synchronization, indicating that motion coupling does not come from changes in air dynamics and requires a physical component that can propagate the kinetic energy generated by the oscillating strips.

To demonstrate the universal synchrony in soft systems, LCN actuators with different sizes were jointed. Owing to the intrinsic mechanical properties, each oscillating actuator possesses a different initial oscillating frequency. Illuminating two jointed LCN actuators with unequal lengths results in the entrainment of the oscillating frequency of the shorter actuator by that of the longer one and eventually both motions become synchronized. Nonetheless, contrasting with rigid systems, the distances at which the two soft actuators are able to communicate are rather short and the synchronization phenomenon ceases when the separation between the two strips reaches 10 mm. Furthermore, the energy transmitted between the soft actuators is low and cannot propagate when the connection between the actuators is rigidified or when only a single strip is stimulated and is thus oscillating. Remarkably, this complex coupling behaviour could be modelled with a reasonable agreement by a standard spring-damper mechanical oscillator combined with a motion model derived for Huygens's experiment on the synchronization of pendulum clocks.

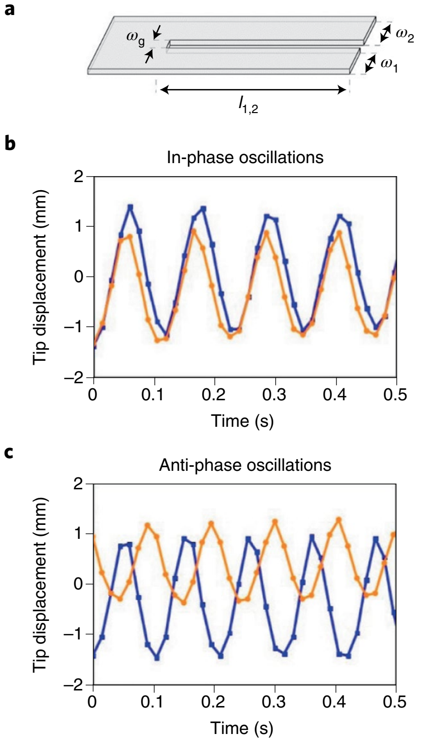

Fig. 1 | Coupled LCN strips with synchronized light-induced oscillations. a, Schematic representation of the coupled oscillators (15 mm ($l_{1,2}$) × 4 mm ($w_1 = w_2$) × 20 µm (thickness)). The strips are separated by a slit of 2 mm (w_g). **b**,**c**, Vertical displacement of the strip tips over time with in-phase synchronized oscillations (**b**) and anti-phase oscillations (**c**). The blue and orange represent the displacement of the individual strips. Figure reproduced with permission from ref. [3], Springer Nature Ltd.

Overall, the results from Vantomme and colleagues confirm that synchronized oscillations share similarities regardless of the nature of the material that compose the actuating system. Synchronization and collective motion are important pathways for the self-organization of living materials. Mimicking such properties in non-living soft materials will amplify molecular variations into macroscopic ones, which may be applicable for developing nano- or microscale sensors or soft robots with lifelike properties. The low energy transfer efficiency from light to mechanical work at the current stage inevitably limits the practical applications of such soft actuators and needs to be addressed in a stepwise manner by material scientists in the future. ❐

Yanlei Yu ✉
Department of Materials Science and State Key Laboratory of Molecular Engineering of Polymers, Fudan University, Shanghai, China.
✉*e-mail: ylyu@fudan.edu.cn*

https://doi.org/10.1038/s41563-021-01159-0

References

1. Bennett, M., Schatz, M. F., Rockwood, H. & Wiesenfeld, K. *Proc. R. Soc. A* **458**, 563–579 (2002).
2. Peña Ramirez, J., Olvera, L. A., Nijmeijer, H. & Alvarez, J. *Sci. Rep.* **6**, 23580 (2016).
3. Vantomme, G. et al. *Nat. Mater.* https://doi.org/10.1038/s41563-021-00931-6 (2021).
4. Pang, X., Lv, J., Zhu, C., Qin, L. & Yu, Y. *Adv. Mater.* **31**, 1904224 (2019).
5. Zhu, C., Lu, Y., Jiang, L. & Yu, Y. *Adv. Funct. Mater.* **31**, 2009835 (2021).
6. Yu, Y., Nakano, M. & Ikeda, T. *Nature* **425**, 145 (2003).
7. White, T. J. & Broer, D. J. *Nat. Mater.* **14**, 1087–1098 (2015).
8. Gelebart, A. H., Vantomme, G., Meijer, E. W. & Broer, D. J. *Adv. Mater.* **29**, 1606712 (2017).

Competing interests

The author declares no competing interests.

PLASMONICS

On-demand emission from Tamm plasmons

Tamm plasmon thermal emitters can provide efficient infrared emission, but are limited by design complexity. Now, the inverse design of Tamm modes facilitated by CdO films on aperiodic dielectric reflectors enables emission with an on-demand spectrum.

Juerg Leuthold and Alexander Dorodnyy

Plasmon polariton enhancements[1–3] — that is, increases in the amplitude of electromagnetic waves due to their coupling with high-density free carriers localized in metals or highly doped materials — are increasingly utilized to improve the efficiency of various photonic devices such as sensors[4], detectors[5], modulators[6] and light sources[7]. One of the simplest strategies to make electromagnetic waves interact with metallic surfaces is to confine them at the interface between the metal and a dielectric layer. The wave is then bound on one side by the metal reflection and on the other side by the total internal reflection of the dielectric. Such electromagnetic surface waves that are strongly coupled to electron oscillations are called surface plasmon polaritons (SPP). However, their reliance on total internal reflection means that SPP modes cannot be directly coupled into from the outside. A versatile alternative is to place a distributed Bragg reflector (DBR) — a periodic stack of dielectric layers with wavelength-selective reflectivity — on top of a metal. This approach enables the formation of Tamm plasmon polaritons (TPPs), which are cavity modes comprising of oscillating electrons and electromagnetic waves. Tamm plasmon polaritons offer customizable characteristics and are capable of light emission enhancement over a broad spectral range. But this approach also adds substantial complexity to the device design. Now, reporting in *Nature Materials*, Mingze He and colleagues show that replacing the metallic layer with doped CdO films provides an additional degree of freedom in the form of a tunable plasma-frequency that, combined with the inverse design of dielectric aperiodic layer stacks, allows the realization of bespoke infrared thermal emitters with on-demand spectrum characteristics based on TPPs[8].

An example of a TPP cavity consisting of a metal and a DBR is depicted in Fig. 1a. The DBR produces a reflection band where complex reflection magnitude and phase are governed by the number of the dielectric layers, layer thicknesses and their dielectric constants, as shown in Fig. 1b. The complex reflectivity of the metal conductor is plotted in Fig. 1c. For a TPP to exist in such a cavity, it must meet the resonance condition of a field travelling back and forth in the cavity. Mathematically, after one roundtrip the complex amplitude will vary by a factor $r_C r_B e^{-2i\phi}$, where $r_C r_B$ is the product of the reflectivity amplitudes at the conductor and the Bragg reflector, respectively, and $e^{-2i\phi}$ represents the phase change per roundtrip in the cavity. Only the wavelengths for which the total accumulated phase is equal to a multiple of 2π will generate sharp cavity resonances, or Tamm modes, as shown in Fig. 1d. The condition for the ideal resonance is $r_C r_B e^{-2i\phi} = 1$ (ref. [9]). The strength of the resonance can be approximated by the quantity $A = |r_C r_B e^{-2i\phi} - 1|^{-1}$, and it is related to the quality of the resonator.

He and colleagues now propose an approach to efficiently design TPPs by improving the control over both complex reflectivities. In their work, they use a doped CdO film instead of a metal and an Al_2O_3/Ge aperiodic layer stack on top of it to form the Tamm modes. They show that by changing the amount of In doping and the carrier concentration thereof, the plasma frequency of CdO can be broadly tuned across the infrared spectral region, facilitating the strong change in the permittivity. Furthermore, they use a stochastic gradient descent approach to optimize the Al_2O_3/Ge stack; this allows them to create cavity modes at a set of given frequencies, reproducing almost arbitrary spectra. Their code allows the design of multimode resonant structures with quality factors of the order of 10 to 1,000. Figure 1e–g depict a triple-band Tamm plasmon cavity with parameters similar to the ones given by He and colleagues. Unlike the conventional DBR that typically has one TPP mode per reflection band, the aperiodic dielectric reflector allows the creation of modes at pre-designed frequencies not limited by the DBR reflection band periodicity.

Experimental verifications by He and co-workers demonstrate the successful fabrication of complex multiband emitter structures and confirm good agreement between simulation and experiment. With such structures, one can now create thermal emitters with an on-demand spectrum. Thermal emitters are frequently used as