A remote control for functional materials

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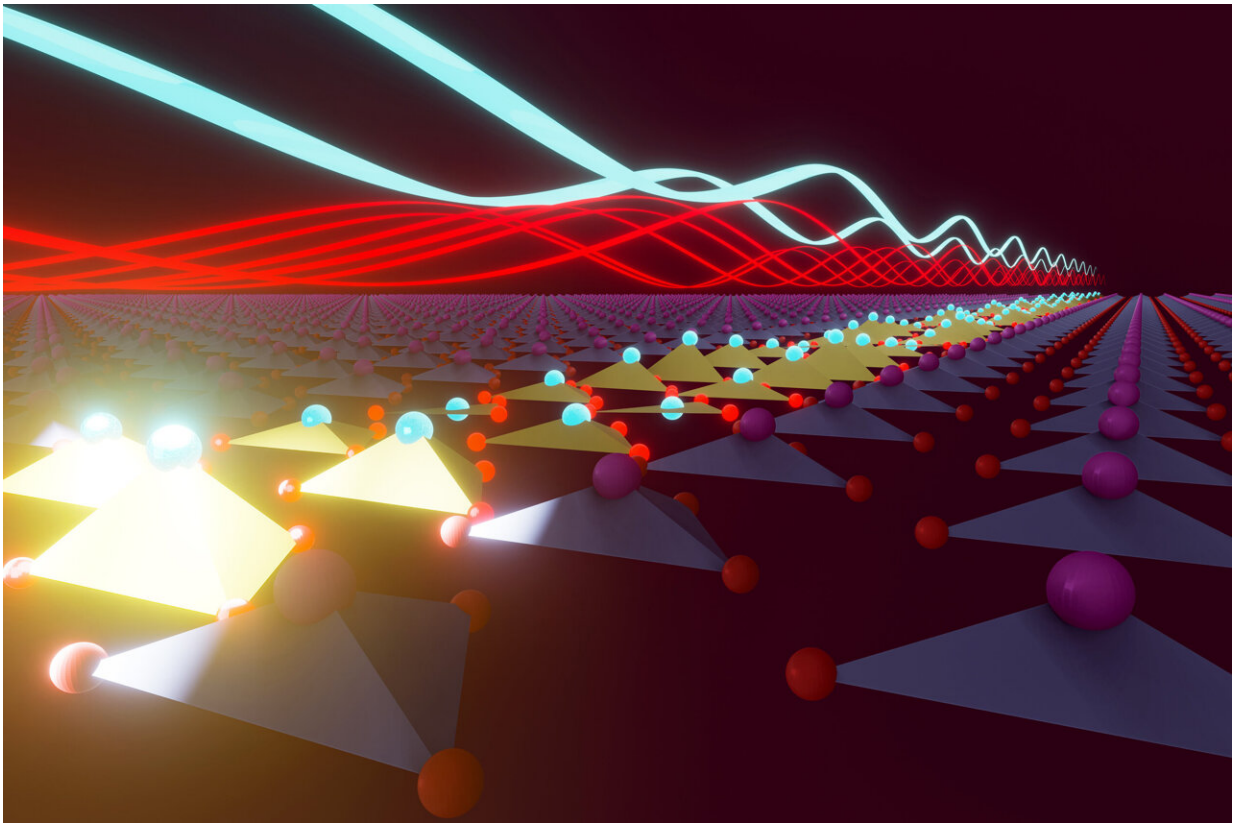

An intense mid-infrared laser pulse hits a ferroelectric LiNbO3 crystal and kicks atomic vibrations only in a short depth below the surface, emphasized by the bright tetrahedra. Through anharmonic coupling, this strong vibration launches a polarization wave, also called polariton, which propagates throughout the remaining depth of the crystal to modulate the ferroelectric polarization. Credit: Joerg M. Harms / MPSD

Intense mid-infrared excitation has been demonstrated as a powerful tool for controlling the magnetic, ferroelectric and superconducting properties of complex materials. Nonlinear phononics is key to this end, as it displaces specific atoms away from their equilibrium positions to manipulate microscopic interactions. So far, this effect has been thought to occur only within the optically excited volume. Now researchers in Hamburg discovered that the polarization reversal in ferroelectric lithium niobate ($LiNbO_3$) even occurs in areas well away from the direct light 'hit'. The hitherto unknown phenomenon—called nonlocal nonlinear phononics—has been published in *Nature Physics*.

Ferroelectric materials such as $LiNbO_3$ possess a static electric polarization generated by lines of positive and negative charge that can be switched with an electric field. This unique property makes these materials the basic building block of many modern electronic components in smartphones, laptops and ultrasound imaging devices. Using laser light to change the ferroelectric polarization is a new approach that allows for extremely fast processes which would be a key step in the development of highly efficient ultrafast optical switches for new devices.

The researchers in Andrea Cavalleri's group at the Max Planck Institute for the Structure and Dynamics (MPSD) used mid-infrared pulses to excite the surface of a $LiNbO_3$ crystal, launching a strong vibration throughout a region that spans a depth of 3 micrometers from the crystal surface. Then, they used a technique called femtosecond stimulated Raman scattering to measure ultrafast changes of the ferroelectric polarization throughout the complete 50 micrometer crystal thickness. The measurements revealed that light pulses with a very high energy density cause the ferroelectric polarization to reverse throughout the entire crystal. By using computational methods to simulate the effects of nonlinear phononics in $LiNbO_3$, the authors found that strong polarization waves called polaritons emerge from the small volume

traversed by the light pulse and move throughout the remaining depth of the crystal. These polariton waves are believed to play a significant role in altering the ferroelectric polarization throughout the sections of the crystal that are untouched by the light pulse.

The results reported by Henstridge et al. add an exciting new piece to the elusive puzzle of ultrafast ferroelectricity, the understanding of which can lead to new device components such as sustainable optical switches. More broadly, this work opens an enormous question concerning whether past and future systems driven by nonlinear phononics can exhibit a similar type of nonlocal character. The ability to manipulate functional properties at a distance could expand the realm of possibilities for incorporating nonlinear phononics into integrated devices and other complex materials, opening new avenues for controlling systems with light.

More information: M. Henstridge et al, Nonlocal nonlinear phononics, *Nature Physics* (2022). DOI: 10.1038/s41567-022-01512-3

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