







# Opportunities in nanoscale probing of laser-driven phase transitions

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For several decades, optical near-field microscopy has facilitated pioneering investigations of photonic excitations at the nanoscale. In recent years, near-field microscopy of terahertz fields has emerged as an important tool for experiments involving phononic and electronic phenomena, rich spatiotemporal dynamics and highly nonlinear processes. Building on this foundation, this Perspective elucidates the transformative opportunities provided by terahertz near-field microscopy to probe ultrafast phase transitions, helping to tackle previously inaccessible challenges of condensed matter physics. Laser-driven phase transitions in many systems are accompanied by the generation of terahertz pulses with spatiotemporal features governed by the complex physics underlying the phase transition. The characterization of these emitted pulses using terahertz near-field microscopy techniques could therefore support the investigation of ultrafast phase transition dynamics. This approach could, for example, allow the observation of ultrafast topological transitions in quantum materials, showcasing its ability to clarify the dynamic processes underlying phase changes.

Recent innovations in time-resolved imaging of electromagnetic fields have provided new methods to investigate phase transitions in new materials with exceptional spatiotemporal resolution. These advances could address emerging challenges in condensed matter physics and material science<sup>1</sup>, especially in strongly correlated, two-dimensional and topological media<sup>2</sup> that involve new types of phase transition that are still not well understood.

Many phase transitions are accompanied by a change in the material's macroscopic electromagnetic properties, arising from a microscopic reorganization of charges or spins inside the material. The intrinsic timescale of such transitions is typically around 0.01–10 ps. This timescale implies that an abruptly triggered phase transition can be accompanied by the emission of pulsed terahertz radiation. Such terahertz emission has been detected in pioneering experiments<sup>3,4,5,6</sup> for various types of phase transition using femtosecond laser pulses as the trigger for the phase transition, yet this phenomenon is not widely appreciated.

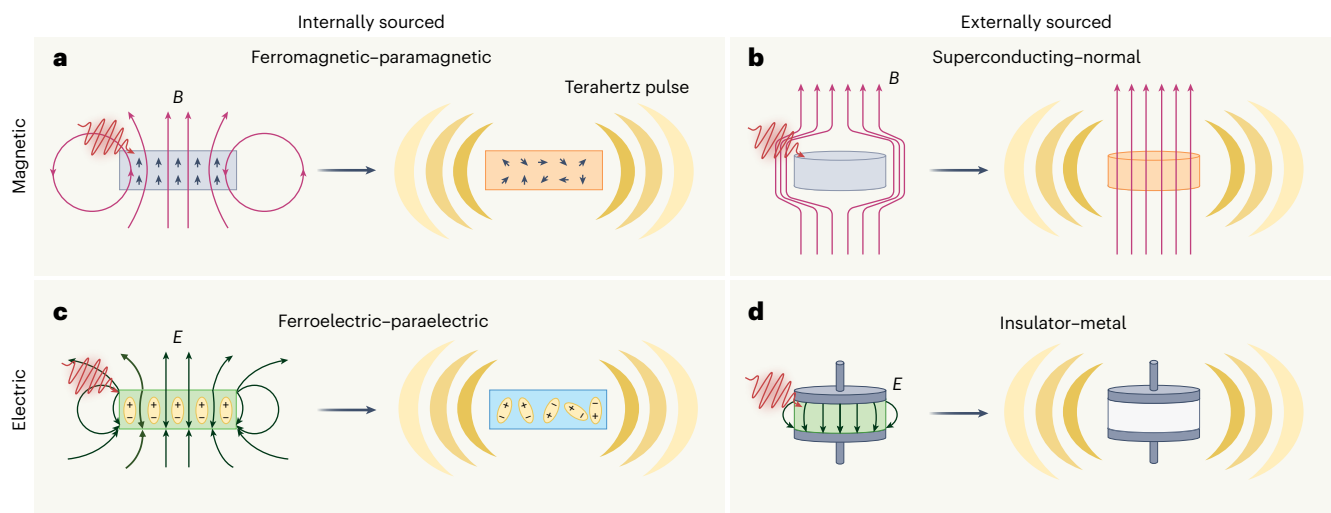
Terahertz emission is universal for a wide range of phase transitions, and looking deeper into it can reveal a general method to probe

the physics of phase transitions in a range of systems. The temporal profile of the emitted terahertz pulse contains information about the internal dynamics of the underlying physical process, commonly probed using terahertz time-domain spectroscopy<sup>7,8</sup>. This technique was applied to the study of various terahertz generation mechanisms in semiconductors, such as optical rectification<sup>9</sup>, shift currents<sup>10,11</sup> and photocurrents<sup>12</sup>.

Recent advances in near-field terahertz measurements provide access to microscopic details on the nanometre scale. Such high resolution could allow the exploration of ultrafast phase transitions in correlated media<sup>2</sup>, ultrafast switching of ferromagnetic<sup>4</sup> and ferroelectric<sup>5</sup> domains (Fig. 1) and ultrafast vortex formation and annihilation in superconductors<sup>13–15</sup>. These phenomena, and many others, could be observed using the associated ultrafast terahertz emission emanating from nanoscale regions.

Here we highlight the prospects of this emerging field, connecting open questions in condensed matter physics with recent advances in nanoscale terahertz imaging and spectroscopy modalities. All phase transitions accompanied by terahertz emission can be loosely

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**Fig. 1 | Emission of an electromagnetic pulse following an abrupt light-induced phase transition.** The terahertz pulse emission (yellow) is a universal phenomenon that results solely from a phase-transition-induced change in some macroscopic electromagnetic property of a material (such as permittivity, permeability, polarization or magnetization). Several manifestations of this phenomenon in different systems are shown, categorized by how their magnetic (purple) or electric (green) fields interact with an incoming laser pulse (red).

**a–d,** Light-induced changes in the magnetic (a,b) or electric (c,d) properties of the material as a result of internally (a,c) or externally (b,d) sourced processes. **a,** Ferromagnetic–paramagnetic transition. **b,** Superconducting–normal transition. **c,** Ferroelectric–paraelectric transition. **d,** Insulator–metal transition. All of these transitions have been reported to occur on (sub)picosecond timescales, corresponding to terahertz emission.

divided into two categories (Fig. 1) based on the source of energy of the emitted pulse: internally sourced or externally sourced.

Internally sourced phase transitions are associated with a change in the material’s spontaneous polarization, either electric or magnetic. In this category, the emission of a terahertz pulse is expected even in the absence of an external d.c. field. The pulse is instead created by abrupt changes and bending of the internal d.c. field. This can happen, for example, when magnetic field lines surrounding a ferromagnet lose their long-range spin order following excitation by a femtosecond laser pulse. The emitted terahertz pulse draws its energy from the material’s internal spontaneous polarization (Fig. 1a).

Externally sourced phase transitions are associated with a change in the material’s electromagnetic response, such as its permittivity, permeability or a nonlinear optical coefficient. In this category, the emission of a terahertz pulse is expected only in the presence of an external d.c. field. This can happen, for example, when magnetic field lines around a superconductor abruptly penetrate a material that lost its diamagnetic response upon heating above its critical temperature  $T_c$  by a femtosecond laser pulse. In this case, the emitted terahertz pulse draws its energy from the external field (Fig. 1b).

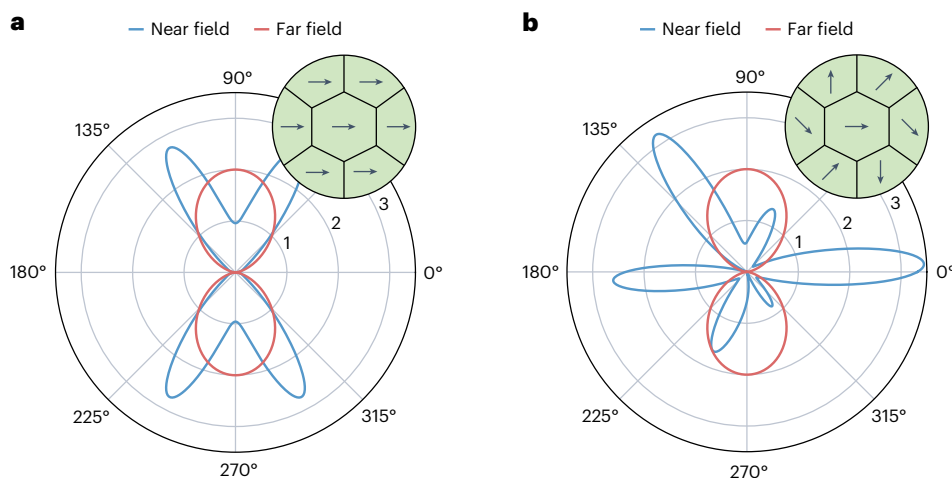
Phase-transition-induced terahertz pulse emission has been experimentally observed in both categories, with notable examples of externally sourced phase transitions including the superconducting phase transition<sup>3,16</sup> and the metal–insulator transition<sup>6</sup>. Observations of internally sourced phase transitions include the ferromagnet–paramagnet transition<sup>4</sup> and the ferroelectric polarization reversal transition<sup>5</sup>. All of these experiments triggered phase transitions using a femtosecond optical excitation.

Although terahertz emission promoted by phase transitions is common to many different physical systems, these experiments remain relatively scarce. Moreover, most experiments register the emitted terahertz pulse in the far field and can therefore uncover only area-averaged information of the macroscopic properties that change during the transition. However, recent advances in time-resolved imaging and spectroscopy experiments have enabled the measurement of terahertz fields in the near field.

When considering a terahertz pulse induced by a phase transition, such measurements could provide valuable spatiotemporal information on the physics underlying the phase transition at the intrinsic time and length scales of the emission process. Figure 2 illustrates this point with simulations of the radiation pattern resulting from a ferroelectric–paraelectric phase transition for two initial domain configurations. Each domain has a well-defined polarization at the start of the experiment, but these dipole moments vanish following pulsed laser excitation, driving a transition to a paraelectric phase. The material imprints its inhomogeneous domain structure on the near field of the emitted terahertz pulse, leading to drastically different near-field angular profiles for different initial domain configurations. The far-field patterns, on the other hand, are dictated by the average polarization (which is the same for both initial conditions), washing out the fine details of the individual domains. The two states are therefore indistinguishable from the far-field signal. For similar reasons, time-resolved measurements in the near field can reveal more details of the different time dynamics of the individual domains.

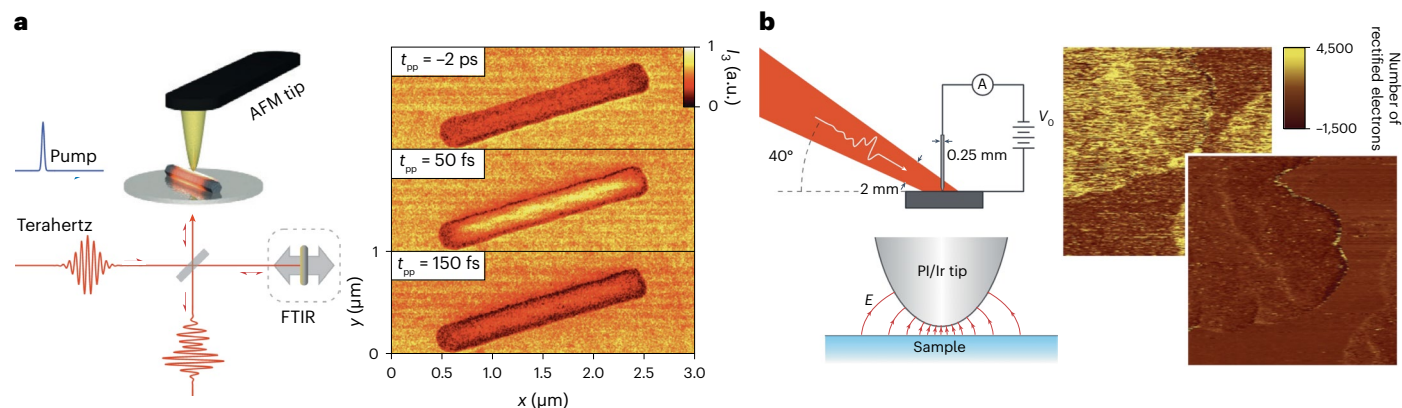
Nanoscale probing of terahertz pulses generated from phase transitions could be experimentally realized using established near-field techniques<sup>17</sup> (Fig. 3). For example, scattering-type scanning near-field optical microscopy<sup>18,19</sup> can record the local terahertz field amplitude scattered off a nanometre-sized tip, thereby overcoming the diffraction limit (Fig. 3a). Laser terahertz emission microscopy<sup>20</sup> relies on an optical femtosecond pulse excitation to generate terahertz fields that scatter off the tip. Alternatively, in terahertz scanning tunnelling microscopy (STM)<sup>21,22</sup>, an optical pump pulse and terahertz probe pulse together induce a transient current in the measuring tip (Fig. 3b). In all of these methods, the tip enables nanometre-scale spatial resolution, giving access to high-momentum processes such as excitation and observations of 2D polaritons confined to nanometre-scale domains<sup>23–25</sup>.

Although tip-based techniques allow access to the local terahertz field emitted through phase transitions at the nanoscale, they are in some sense disruptive, as the presence of the tip inevitably alters the near field it helps to observe. The ultrafast transmission electron microscope (UTEM) has emerged as a promising complimentary platform



**Fig. 2 | Comparing near-field and far-field terahertz emission experiments.** **a,b**, Simulated angle-resolved near-field (blue) and far-field (red) radiation patterns emitted following a ferroelectric–paraelectric phase transition for aligned **(a)** and misaligned **(b)** initial ferroelectric domain configurations

(green). The near-field angular profiles capture the initial microscopic domain structure, showing how it differs between the two configurations, whereas the far-field profiles are identical in both scenarios.



**Fig. 3 | Tip-based experiments for time-resolved nanoscale imaging of terahertz fields.** **a** Example of scattering-type near-field scanning optical microscopy. Left: an InAs nanowire is excited by a near-infrared pump pulse and probed by a terahertz probe pulse. The nanowire’s near fields are scattered by a nanometre-sized tip, which can then be recorded using various methods, such as Fourier transform infrared spectroscopy (FTIR). Right: the terahertz electric near-field intensity distribution  $I_3$  at pump–probe delay times  $t_{pp}$ , showing the spatiotemporal dynamics of the photoinjected free carriers in the InAs

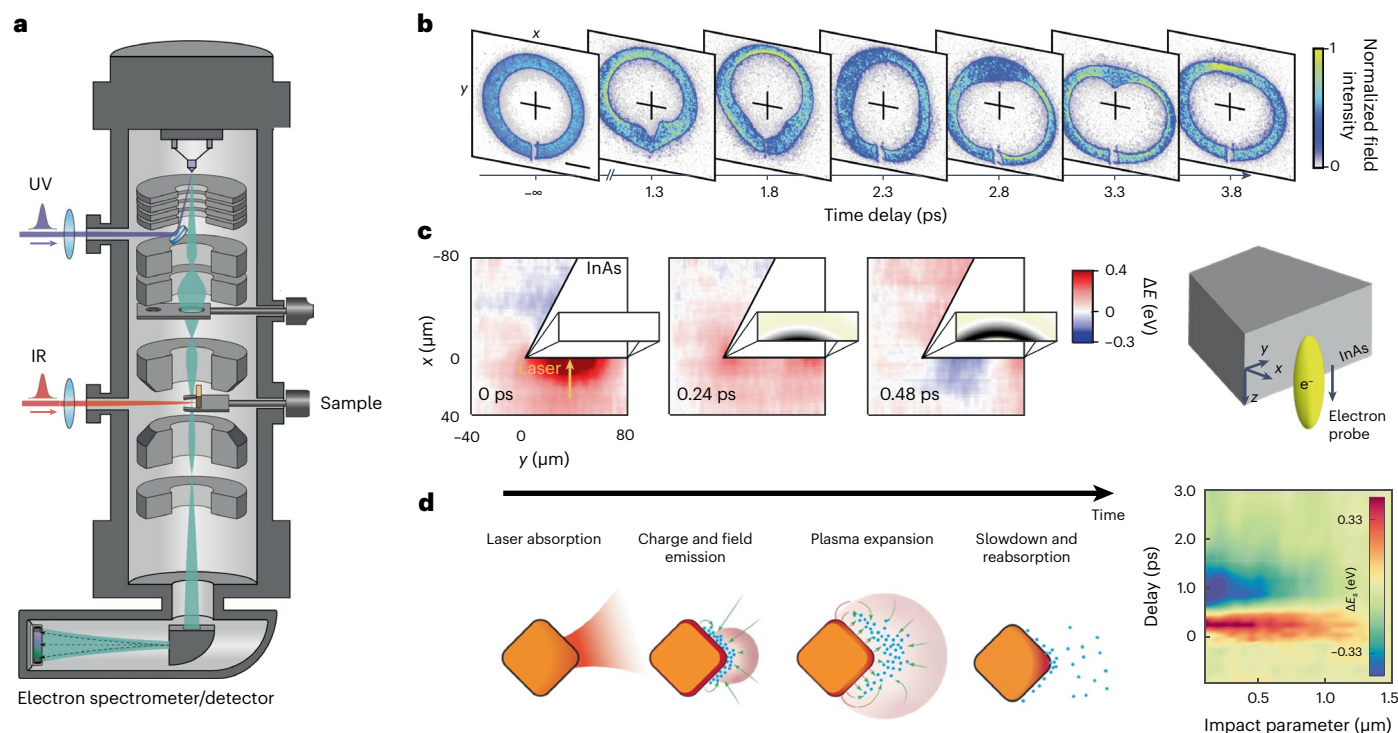
nanowire<sup>18</sup>. **b**, Example of terahertz-pulse-induced tunnelling in an STM. Left: two gold nano islands on a highly ordered pyrolytic graphite surface are excited by a near-infrared pump pulse and probed by detecting the emitted terahertz electric near field with a voltage-biased Pt/Ir STM tip wire. Right: terahertz STM images record the terahertz pulse autocorrelation at time lags of  $\sim 500$  fs (left) and 0 fs (right)<sup>21</sup>. Figure adapted with permission from: **a**, ref. 18, Springer Nature Limited; **b**, ref. 21, Springer Nature Limited.

for non-disruptive probing of the spatiotemporal dynamics of phase transitions with combined nanometre and femtosecond resolution, using free electrons to measure the near field in a direct manner<sup>26</sup>. These microscopes employ femtosecond laser pulses to excite the sample, synchronized with femtosecond free-electron pulses that probe the resulting terahertz near fields (Fig. 4a). Measuring the terahertz near field facilitates the reconstruction of the underlying charge or spin dynamics, and thus of the phase transition dynamics in the material.

Such measurements can be realized in multiple ways. Ultrafast electron deflectometry<sup>27</sup> (Fig. 4b) measures electrons that change their transverse momentum after elastic scattering off the emitted terahertz field, leading to a distorted image of the studied material in the detector plane, from which the local field amplitude is extracted. Alternatively, charge dynamics electron microscopy<sup>28,29</sup> (Fig. 4c,d) relies on the energy change of the probing free electrons following their inelastic scattering off a terahertz field. This energy change results from the interaction of each electron with the longitudinal

component of the electric field along the electron’s path, from which the terahertz near field is reconstructed. Important steps towards ultrafast electron holography are also being made, with the prospect of detecting subtle changes in the electron phase via point-projection microscopy<sup>30</sup>, off-axis holography<sup>31</sup> and Lorentz transmission electron microscopy<sup>32,33</sup>. Ultrafast electron microscopes also support other techniques for nanoscale imaging of ultrafast phase transitions, not via their accompanying terahertz emission, but through ultrafast electron diffraction<sup>34</sup> or dark-field microscopy<sup>35</sup>.

Electron-based techniques are fundamentally different from tip-based techniques. An electron measures the near field in a direct and non-invasive manner, unlike tip-based techniques, which inevitably disturb the near field. Electron-based techniques can potentially extract three-dimensional information via tomographic angle-dependent measurements, and are compatible with cryogenic temperatures as the instruments lack moving mechanical parts. However, because of energy–momentum conservation, electrons can only couple to



**Fig. 4 | Free-electron-based experiments for time-resolved nanoscale imaging of terahertz emission.** **a**, Illustration of a UTEM. An ultraviolet (UV) pulse is guided to the TEM cathode, generating femtosecond electron pulses, synchronized with an additional infrared (IR) pulse that pumps the sample. The electron pulse travels down the TEM column, probing the emitted near field inside or near the sample. The post-interaction electron energy is measured using an electron energy spectrometer. The UTEM enables a range of imaging methods of terahertz near fields. **b**, Terahertz waveforms at multiple time delays imaged using ultrafast electron deflectometry<sup>27</sup>. The scale bar in the leftmost image is 150  $\mu\text{m}$ . The field intensity is normalized to the maximum value. **c, d**, Charge dynamics electron microscopy based on terahertz-mediated inelastic electron scattering is used to probe the electron energy change  $\Delta E$  for multiple time delays and electron beam positions. Using this technique, it is possible to

image the terahertz near field and extract the electron–hole transport in a bulk semiconductor<sup>29</sup> (**c**) and nanoscale plasma dynamics in a vacuum<sup>28</sup> (**d**). **c**, Left: Spatiotemporal maps of the electron energy change  $\Delta E$  after traversing near an InAs sample, from which the charge distribution on the sample is reconstructed (as depicted in the Insets). Right: Illustration of an electron probe beam traversing near an InAs sample. **d**, Left: Illustrations depicting the distinct stages of plasma dynamics following laser irradiation of a solid target. Right: Measured spatiotemporal variation of the electron energy change  $\Delta E$  after interacting with the plasma cloud. These approaches could similarly probe terahertz signals arising from ultrafast phase transitions. Figure adapted with permission from: **a, c**, ref. 29, American Chemical Society; **b**, ref. 27, AAAS; **d**, ref. 28 under a Creative Commons license CC BY 4.0.

fields with a specific momentum component, whereas the extreme momentum mismatch in tip-based technologies supports both in- and out-coupling of high-momentum field components.

In terms of resolution, the two approaches are comparable at present. Tip-based techniques can achieve few-nanometre spatial resolution, determined by the tip geometry and scanning parameters, while also supporting temporal resolutions between tens and hundreds of femtoseconds<sup>18,21</sup>. In electron-based techniques, a spatiotemporal resolution of tens of nanometres and hundreds of femtoseconds has been reported<sup>26</sup>. However, both the spatial and temporal resolution achievable with electron-based techniques are probably going to substantially improve in the coming years once limiting factors have been mitigated. Improvement strategies could include the introduction of electron optics adapted for ultrafast electron microscopes, which could achieve lower space–charge effects and allow us to acquire a better understanding of pulsed-electron emittance trade-offs.

One can extract physical insight into phase transitions from emitted terahertz near fields using several methodologies. Time-resolved terahertz emission spectroscopy can reveal allowed absorption or emission channels, which are associated with various excitations governed by symmetry arguments and selection rules<sup>36</sup>. The terahertz transient pulse profile itself could also provide valuable information on, for example, the propagation of the phase transition inside a material. The local terahertz field map, in conjunction with Maxwell’s equations, could facilitate the reconstruction of the underlying charge or

spin dynamics—either directly or by solving the inverse problem. The direct solution requires complete knowledge of the spatiotemporal vector-field distribution. Solving the inverse problem can be accomplished by applying an appropriate condensed matter model to the studied physical system<sup>28,29</sup>. A sequence of measurements carried out while changing the experimental conditions (pump intensity, wavelength and so on) could then be used to retrieve the model parameters.

Besides phase transitions, a many other phenomena can lead to terahertz emission (Fig. 4c,d). Regardless of the method used, the measured signal arising from the emitted terahertz radiation should be carefully investigated to distinguish the different contributions. As the physics of phase transitions is usually highly nonlinear, there is merit in using intense and ultrashort laser pulses (typically tens of femtoseconds and occasionally few femtoseconds) to drive the phase transition. Using mode-selective excitation<sup>37</sup>, in which short pump pulses at optical frequencies target specific resonances in materials, has the added benefit of reducing the heat load on the studied sample. Furthermore, ultrashort pump pulses enable the temporal separation of the dynamics of interest from the pump pulse itself, thus reducing potential artefacts from the interaction of the pump with the probe.

Nanoscale probing of ultrafast phase transitions is especially attractive for research of metastable or transient phases—hidden phases that are only revealed by pulsed excitation, such as light-induced superconductivity<sup>38</sup>, optically driven ferroelectric phases (as in  $\text{SrTiO}_3$ <sup>39,40</sup>) and extreme optical nonlinearities in solids that facilitate

high-harmonic generation processes<sup>41,42</sup>. Similarly, it would be interesting to explore how symmetry-related properties break on ultrafast timescales<sup>43</sup>. The high spatial resolution is of particular relevance to phase transitions in topological edge states<sup>44–47</sup> and in low-dimensional systems such as interfacial currents in transition metal dichalcogenide heterostructures<sup>48</sup>. So far, most of these phenomena have been probed with area-averaging techniques. A near-field approach could provide new insight into yet-to-be-explored mechanisms responsible for the phase transitions such as the underlying quasiparticle dynamics<sup>49</sup>. We expect nanoscale measurements of terahertz pulses triggered by phase transitions to become standard practice in the investigation of ultrafast phenomena in quantum materials.

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## Competing interests

The authors declare no competing interests.

## Additional information

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