Research Highlights from Ultrafast and Terahertz spectroscopy (UFTS) group led by Dr. N. Kamaraju in collaboration with Prof. Thiruppathaiah Setti's group from SNBNCBS Kolkata (Single crystals), Prof. Shreeganesh Prabhu's group in TIFR Mumbai, and Prof. Sidhartha Lal's group in Department of Physical Sciences, IISER Kolkata.

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In this work, we have taken a novel and unexplored route of using picosecond strain pulses towards understanding the phenomenon of magnetic dimensional crossover (MDC) in a two-dimensional (2D) van der Waals Heisenberg ferromagnet $CrSiTe_3$ using femtosecond time resolved experiments for the first time to the best of our knowledge.

Unravelling the transition from short-range 2D intraplanar magnetic ordering (SRMO) to long-range 3D interplane magnetic ordering in two-dimensional van der Waals materials carries significant potential for both fundamental scientific exploration and technological innovation. However, direct observation of the intricate stages of this magnetic dimensional crossover (MDC) remains a significant experimental challenge. Employing the magneto-elastic coupling for this offers a promising approach, but detecting the subtle lattice response to short-range magnetic order (SRMO) remains challenging. While recent studies using second harmonic generation have hinted at a two-step MDC process, a definitive time-resolved observation has proven elusive.

We adopt a novel methodology, utilizing picosecond acoustic strain pulses induced by femtosecond lasers in $CrSiTe_3$, to scrutinize the microscopic mechanisms underpinning MDC in the time domain. This groundbreaking technique enables us to witness firsthand the nuanced yet pivotal impact of spin fluctuations and ordering on the lattice, thereby unveiling the progression of magnetic order with unparalleled precision. Our examination of the temporal and spectral responses of the strain pulse across a diverse temperature range provides a comprehensive depiction of the MDC process. Our investigation further suggests that the generation of strain is influenced not only by the thermoelastic and deformation potential origins, but also by the magnetic strain inherent in the ferromagnetic phase. Consequently, we not only discern a distinct signature of the multi-stage MDC pathway but also uncover hitherto unobserved facets of the interaction between spin dynamics and lattice vibrations.

The renormalization in the strain spectrum within the frequency domain, along with the phase change in the time domain, is encapsulated in a phenomenological theoretical model of the spin-lattice interaction, which we have delineated in the manuscript. Furthermore, the ultrafast carrier dynamics that contribute to the electronic multi-exponential background also exhibit signatures of MDC. These discoveries not only facilitate a more profound comprehension of magnetic ordering in layered 3D materials, but also herald a new era of time-resolved experiments with the potential to revolutionize our understanding of spin-based phenomena. We believe the techniques and findings presented in this study could be used as a basis for further research into 2D van der Waals materials and their magnetic properties in designing a new generation of spin-based optoelectronic devices.

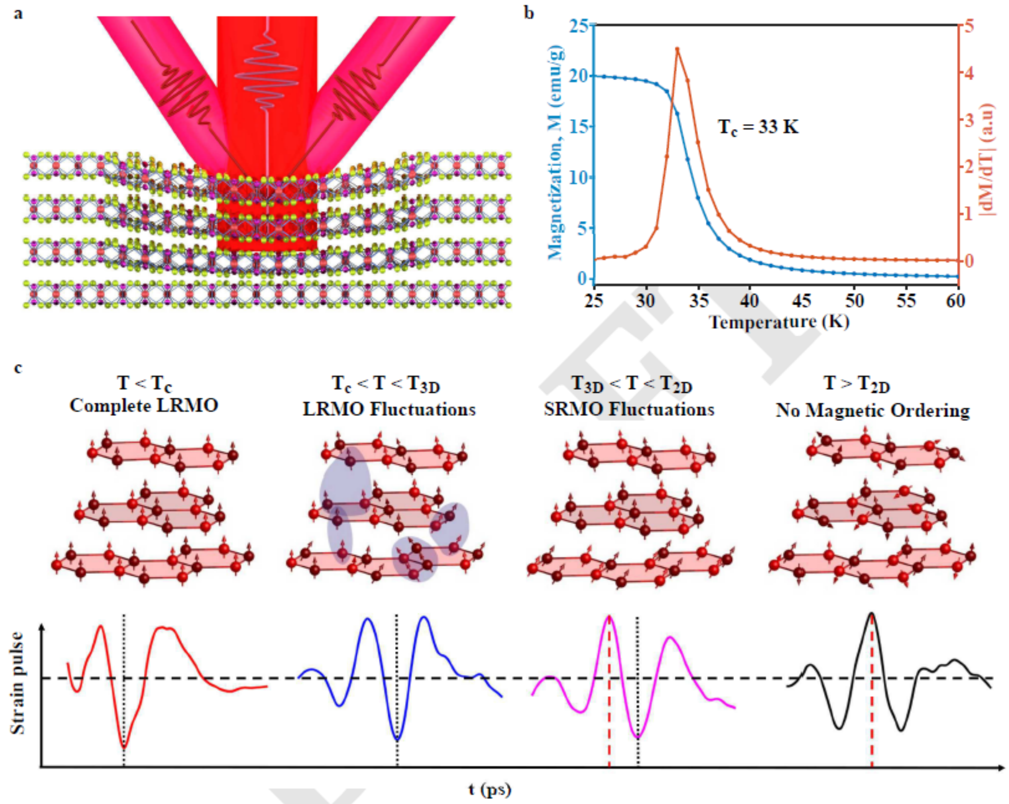

Fig. 1. Detection of dimensional crossover using strain pulses: (a) Schematic of ultrafast pump probe technique to generate and detect the strain pulse (b) Variation of magnetization as a function of temperature (blue filled hexagons are the data and the blue lines are the guide to the eye) and absolute value of derivative of magnetization (dM/dT) versus temperature (orange lines are the guide to the eye, connecting the orange filled hexagons(data)). Transition temperature is found here to be 33 K, (c) The various stages of magnetic ordering captured by the strain pulse, depicted along with the schematic of 'Cr' spin ordering in the layers. One can notice that the maximum of the strain pulse is negative at $T < T_c$ and the shape of the strain pulses gradually changes to a stage where the maximum of the pulse becomes positive at $T > T_{2D}$. The horizontal black dashed line denotes the reference level (y = 0). Vertical lines, distinguished by dotted and dashed styles, identify the extrema of the strain pulses. The red dashed line marks the tensile strain (positive peak), while the black dotted line marks the compressive strain (negative peak). Note that at $T > T_{2D}$, the maxima and minima are comparable and hence both extremas are marked.

Credits: High quality single crystals were provided by Prof. Thirupathaiah Setti's group from SNBNCBS Kolkata (https://people.bose.res.in/faculty/fac_new/setti.html), the experiments were done in my group at DPS, IISER Kolkata and also in Prof. Shriganesh Prabhu's group in TIFR (https://sites.google.com/.../thzf.../people/prof-shriganesh-Shriganesh Prabhu), Mumbai. The phenomenological theory was provided by Prof. Siddhartha Lal's group in DPS, IISER Kolkata (https://epqm.github.io/).