

Spatially resolved ultrafast magnetic dynamics launched at a complex oxide hetero-interface

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Static strain in complex oxide heterostructures^{1,2} has been extensively used to engineer electronic and magnetic properties at equilibrium³. In the same spirit, deformations of the crystal lattice with light may be used to achieve functional control across hetero-interfaces dynamically⁴. Here, by exciting large amplitude infrared-active vibrations in a LaAlO₃ substrate we induce magnetic order melting in a NdNiO₃ film across a hetero-interface. Femtosecond resonant soft x-ray diffraction is used to determine the spatiotemporal evolution of the magnetic disordering. We observe a magnetic melt front that propagates from the substrate interface into the film, at a speed that suggests electronically driven motion. Light control and ultrafast phase front propagation at hetero-interfaces may lead to new opportunities in optomagnetism, for example by driving domain wall motion to transport information across suitably designed devices.

In transition metal oxides, rearrangements in electronic and magnetic properties can be triggered by the application of magnetic⁵ and electric fields⁶, or pressure^{7,8}. Switching has also been demonstrated in these materials using femtosecond optical excitation, at near-visible^{9,10,11,12}, mid-infrared^{13,14,15,16}, or THz^{17,18,19} wavelengths. Recently, selective excitation of lattice modes in the mid-infrared has been applied to complex oxide heterostructures, where the functional material can be separated from the optically excited region. By directly driving the LaAlO₃ substrate infrared-active modes in a LaAlO₃/NdNiO₃ heterostructure, an insulator-metal transition was triggered across the interface in the nickelate film⁴.

Here, we apply time-resolved resonant soft x-ray diffraction (RSXD) to measure the concomitant magnetic response in the NdNiO₃ film, with nanometer spatial and femtosecond temporal resolution. We find evidence of inhomogeneous magnetic melting dynamics, with a melt front that propagates from the interface into the NdNiO₃ film at speeds of order of, or even in excess of, the speed of sound. An insulator-metal transition initiated at the hetero-interface and propagating inward, possibly locked to a wave of local octahedral distortions, are considered to explain these observations. Theoretical calculations using a model Hamiltonian indicate that itinerant charge carriers in a spin-ordered correlated electron insulator can dynamically scramble the magnetic order efficiently as they move into the film.

At about 200 K, metallic paramagnetic NdNiO₃ undergoes a transition into a low-temperature antiferromagnetic insulating state²⁰. This electronic and magnetic phase transition is concomitant with a structural transformation from an orthorhombic (Pbnm) to a monoclinic (P21/n) crystal structure. The transition

temperature depends on epitaxial strain²¹, demonstrating sensitivity to lattice distortions.

We study a compressively strained 30 nm thick NdNiO₃ film grown on a (111) LaAlO₃ substrate (see Methods). The low-temperature antiferromagnetic order on the Ni and Nd sublattices, shown schematically in Fig. 1(a), can be observed by RSXD at the pseudo-cubic (1/4 1/4 1/4) wave vector²². Figure 1(b) shows the static photon-energy dependent magnetic diffraction from this ordering, measured at the Ni L_{2,3} edges using synchrotron radiation.

In a femtosecond RSXD experiment at a free electron laser (FEL), the sample was excited by mid-infrared pulses resonant with the highest-frequency LaAlO₃ substrate phonon, which is well separated in energy from equivalent phonons of the NdNiO₃ film (see Figure 1(c); further experimental details are given in the Methods). Figure 1(d) shows the vibrationally induced intensity changes of the (1/4 1/4 1/4) diffraction peak, which reflect the dynamics of the antiferromagnetic order in the NdNiO₃ film. The peak intensity dropped by about 80 % within 1.6 ps, and recovered on the tens of picosecond time scale. In contrast to the case of direct interband excitation, for which the antiferromagnetic peak dropped promptly²⁴, the 1.6 ps time constant for the initial reduction was significantly longer than the 200-fs pump pulse duration and the 250-fs time resolution of the experiment, where the latter is limited by the jitter between the FEL and the optical laser. We compare these dynamics to the time needed for the film to become metallic, as measured by the transient reflectivity in the 1–5 THz range induced by the same mid-infrared excitation (green dots in Fig. 1(d)).⁴ These two similar timescales, which reflect only

average changes over the whole film, suggest a connection between the insulator-to-metal transition and the melting of magnetic order.

In Figure 2, we plot the transient θ - 2θ scans for the $(1/4 \ 1/4 \ 1/4)$ diffraction peak, sensitive to the out-of-plane antiferromagnetic ordering. In equilibrium, i.e. at negative time delay, a narrow diffraction peak and Laue oscillations are observed, indicating that magnetic order is homogeneous across the entire film, with sharp magnetic boundaries.

Figure 2 further shows that a significant peak broadening and a suppression of the Laue oscillations accompany the photo-induced reduction in peak intensity. The broadening of the diffraction peak implies that the excitation melts the magnetic order only over a fraction of the film. The suppression of the Laue oscillations indicates that the boundary between the ordered and disordered regions of the film is not sharp.

Note that throughout these dynamics the in-plane correlation length, as measured by transverse rocking curves (θ scans), remains unchanged (see Supplementary Information). Hence, the dynamics discussed here are one-dimensional, evolving along the sample growth direction.

The spatial distribution of the magnetic order at a time delay τ was analyzed quantitatively with the following expression for kinematic diffraction

$$I_{\tau}(q) \propto \left| \int_0^D F(z, \tau) e^{-iqz} dz \right|^2. \quad (1)$$

Here, the magnetic profile is represented by the space- and time-dependent structure factor $F(z, \tau)$, where $q = 4\pi \sin \theta / \lambda$ is the magnitude of the scattering wave

vector (with θ the diffraction angle and λ the x-ray wavelength), z the distance into the film, and D the film thickness.

Previous work has shown that these complex dynamics are initiated at the buried interface⁴. Also, the transient θ - 2θ scans show that the fraction of material, which is melted, increases with time, and that the order-disorder interface becomes progressively smeared in time. Hence, we chose a functional form $F(z, \tau)$ that describes a soliton-like demagnetization front, propagating from the hetero-interface into the thin film with a smeared and adjustable phase front:

$$F(z, \tau) = F_0 \cdot \left(\frac{1}{2} + \frac{1}{2} \operatorname{erf}\left(\frac{z - z_f(\tau)}{d_f(\tau)}\right) \right). \quad (2)$$

In this case, F_0 is the equilibrium structure factor, and z_f and d_f are the time-dependent position and width of that phase front separating the unperturbed antiferromagnetic order from the disordered region of the film. Numerical fits of Eqs. (1) and (2) to the diffraction peak at selected time delays are shown as red solid lines in the lower panel of Fig. 2 and give excellent agreement with the data.

Figure 3(b) shows the corresponding early time scale evolution of the space-dependent magnetic order parameter, represented as $|F(z, \tau)|^2$. At negative time delays, the NdNiO₃ film is homogeneously ordered, with a sharp boundary at the hetero-interface. The mid-infrared excitation induces heterogeneous melting, with a demagnetizing phase front that propagates halfway into the film – before coming to a stop at ~ 2 ps time delay. This phase front leaves a magnetically disordered region behind. The time-dependent position of the phase front z_f is plotted in Figure 3(c),

which suggests that the magnetic melt front propagates at a speed of order of, or even faster than the longitudinal sound velocity measured in a NdNiO₃ film²³.

The re-magnetization dynamics are shown in Figure 4. About 10 ps after excitation, the demagnetized part of the film starts recovering. Here, the phase front begins shifting back towards the substrate interface, and at the same time the domain boundary broadens further.

The heterogeneous dynamics discussed above are unique to the mid-infrared excitation of the substrate lattice. Near-infrared illumination of the same heterostructure at 800 nm wavelength, which involves significant homogeneous charge excitation in the nickelate film²⁴, shows that in this case the magnetic order is uniformly melted over the entire film. This can be clearly deduced from Figs. 5(a) and (b) unveiling that the 800-nm excitation reduces the intensity of the magnetic (1/4 1/4 1/4) diffraction peak and of the Laue oscillations, without any noticeable change in the peak width. An analogous fitting procedure as applied above for the mid-infrared substrate excitation (see also the red solid lines in Figure 5(b)), shows the uniform and instantaneous decrease of the magnetic order parameter across the whole film thickness.

We next turn to a discussion of the possible physical mechanisms underlying these observations. Firstly, we can exclude direct absorption of the mid-IR pump pulse in the nickelate film as being responsible for the observed magnetization dynamics. This is not only because the energy deposited in the nickelate is negligible (3% of the incident energy), but also because the 1- μm penetration depth in NdNiO₃ would imply homogeneous excitation. Note further that only a uniform decrease of the

magnetic order is observed when heating the NdNiO₃ film up to the phase transition temperature. Secondly, as the speed of the magnetic melt front is as high or higher than the nominal longitudinal sound velocity²³, this cannot be caused by heat propagation, which is significantly slower. As a third possibility, one could consider a structural phase transition triggered at the buried interface, propagating inward and driving the magnetic melt front. However, a structural phase front propagates far slower than the speed of sound and is therefore also an unlikely explanation²⁵. Finally, a propagating acoustic wave is also unlikely, as it is not clear how the resonant excitation of an optical phonon could launch a propagating strain pulse on this short time scale. Note also that the strain extracted from the shift in the diffraction peak is never larger than 10⁻³.

We propose that the direct excitation of the infrared-active substrate phonon mode induces octahedral distortions^{26,27,28} across the interface, to locally act on the electronic and magnetic ordering of the nickelate film. Indeed, similar scenarios have been predicted for perovskite heterostructures in the static case²⁹. Propagation of the phase would then presumably be driven by electronic rather than magnetic effects. We note that the anisotropic magnetic interaction stabilizing the Neel order introduce a spin gap and hence flatten the dispersion of magnons (see Supplementary Information for details), resulting in reduced group velocity and magnon localization.

We believe that charge carriers, which are made mobile at the interface and are very efficient at randomizing spin correlations³⁰, are more likely to drive the magnetic front. As these itinerant carriers acquire kinetic energies that exceed magnetic

energy scales, these can sustain a front that propagates. The scrambling of antiferromagnetic order would transfer initial kinetic energy into the magnetic sector³¹, eventually leading to a stalling of the phase front. Although a mechanistic description of this process requires materials specific calculations, which are beyond the scope of the present paper, a model Hamiltonian description, in which charges are freed at the interface and are made mobile and diffuse into the film, confirms these qualitative arguments (see Supplementary Information). This scenario would also be compatible with the similar time scales observed for the insulator-metal transition and the magnetic order melting presented in Fig. 1(d).

In summary, we have shown that the dynamics of magnetic disordering in complex oxide heterostructures follows distinctly new physical pathways when the substrate lattice is excited. We demonstrate that magnetic melting is initiated at the buried interface and propagates into the film at speeds of order of or higher than the speed of sound. We assign the underlying physics to an electronic itinerancy front, which carries spin disordering and possibly local lattice distortions. The ability to control such ultrafast magnetic phase fronts with light may be conducive to new applications in optomagnetic devices, in which information may be encoded and shuttled in domain walls at faster rates than more established spin torque driven walls³².

METHODS

The NdNiO₃ thin film was deposited on the (111) LaAlO₃ single crystal substrate by off-axis RF magnetron sputtering³³. This substrate provides 0.5% compressive strain to the material and reduces the metal-insulator-transition temperature from a bulk value of 200 K to about 130 K for the chosen film thickness.

The static RSXD data, presented in Fig. 1(b), were taken at the I06 beamline of the Diamond Light Source synchrotron x-ray source.

Femtosecond RSXD experiments were carried out at the SXR beamline of the Linac Coherent Light Source free electron laser³⁴. The excitation pulses of 200 fs duration were tuned to 15 μm wavelength (82 meV photon energy) and focused onto the sample with a fluence of 4 mJ/cm². The sample was mounted onto an in-vacuum diffractometer³⁵ and cooled to 40 K, i.e. into the antiferromagnetic insulating state. The FEL, which operated at 120 Hz repetition rate, was tuned to the 852 eV Ni L₃ edge. The bandwidth of the x-ray pulses was reduced to below 1 eV by a grating monochromator. Diffracted x-rays were detected as function of the time delay relative to the mid-infrared excitation pulses. An avalanche photodiode enabled pulse-to-pulse normalization of the diffracted to the incident light intensity.

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AUTHOR CONTRIBUTIONS

A.D.C., M.F. and A.C. conceived this project. M.F., A.D.C., R.M., V.K., S.B.W., S.S.D., and J.P.H performed the experiment at the LCLS, supported by W.F.S., J.J.T. and G.L.D (beamline), M.P.M. and J.R. (laser), Y.D.C and W.S.L. (experimental endstation). The sample was grown by R.S., P.Z., and J.-M.T.. M.F. and A.D.C. analyzed the data with help from H.B.. S.R.C. and D.J. provided the model Hamiltonian theory. M.F., A.D.C. and A.C. wrote the manuscript, with feedback from all co-authors.

FIGURE CAPTIONS

Fig. 1. Magnetic ordering in NdNiO₃ and the vibrationally induced phase transition. **a** Visualization of the complex antiferromagnetic ordering in NdNiO₃. **b** Energy scan of the (1/4 1/4 1/4) diffraction peak, associated with the magnetic order on the Ni sub-lattice, around the Ni L_{2,3} edges. These data were measured on the NdNiO₃/LaAlO₃ heterostructure at 15 K temperature. **c** Wavelength dependent inverse penetration depth of mid-infrared light around the NdNiO₃ (green) and LaAlO₃ (blue) high-frequency phonon resonances. The energy spectrum of the excitation pulses at 15 μm wavelength (black data points, fitted by a Gaussian profile in red) is tuned into resonance with the substrate mode. **d** Blue: Changes of the peak intensity of the Ni L₃ diffraction peak at 852 eV, following this optical excitation with a fluence of 4 mJ/cm². The green data points show the transient THz reflectivity of the same sample under identical excitation conditions. The melting of the magnetic order and the insulator-metal transition both take place on the same time scale.

Fig. 2: Temporal evolution of the antiferromagnetic order in reciprocal space. **a** Lattice-induced dynamics of the momentum dependence of the (1/4 1/4 1/4) Ni L₃-edge diffraction peak (θ - 2θ scans), on both short and long time scales. The diffraction intensity is color-coded on a logarithmic scale. **b** The same diffraction peak at selected time delays before and after the mid-infrared excitation (blue dots), overlaid with numerical fits (red lines) that assume a heterogeneous melting of the antiferromagnetic order triggered at the NdNiO₃/LaAlO₃ interface as described in the text.

Fig. 3: Real space dynamics of the antiferromagnetic order. **a** Schematic illustration of the demagnetization process. At negative time delay, the NdNiO₃ film is homogeneously ordered. Direct excitation of a substrate phonon triggers a magnetic melt front, propagating from the interface into the film. **b** Short time scale spatial dynamics of the magnetic order parameter along the [111] direction, extracted from the numerical fits to the diffraction peaks shown in Figure 2. **c** Centre position of the phase front, separating the melted from the unperturbed region of the film (blue dots). The error bars are the standard errors obtained from the fits. The red solid line is a guide to the eye. The striped area marks the sonic regime, given by the 4.1×10^3 m/s NdNiO₃ longitudinal speed of sound extracted from acoustic echo times in Ref. 23.

Fig. 4: Recovery dynamics of the antiferromagnetic order. **a** Long time scale spatial dynamics of the magnetic order parameter along the [111] direction, extracted from the numerical fits to the diffraction peaks (shown in Fig. 2) at various time delays. **b** Time-dependent position and width of the magnetic phase front corresponding to the data shown in panel (a). The error bars are the standard errors obtained from the fits to the diffraction peaks (shown in Figure 2).

Fig. 5: Magnetization dynamics following Ni charge transfer excitation at 800 nm. **a** Photo-induced dynamics of the $(1/4 \ 1/4 \ 1/4)$ Ni L₃-edge diffraction peak in reciprocal space, with the diffraction intensity color-coded on a logarithmic scale. The excitation fluence is about 4 mJ/cm². **b** The same diffraction peak at selected time delays before and after the near-infrared excitation (blue dots), together with numerical fits (red solid lines) as described in the text. **c** Spatial dynamics of the

magnetic order parameter along the [111] direction (extracted from these fits),
showing homogeneous photo-induced demagnetization.

FIGURES

Fig.1

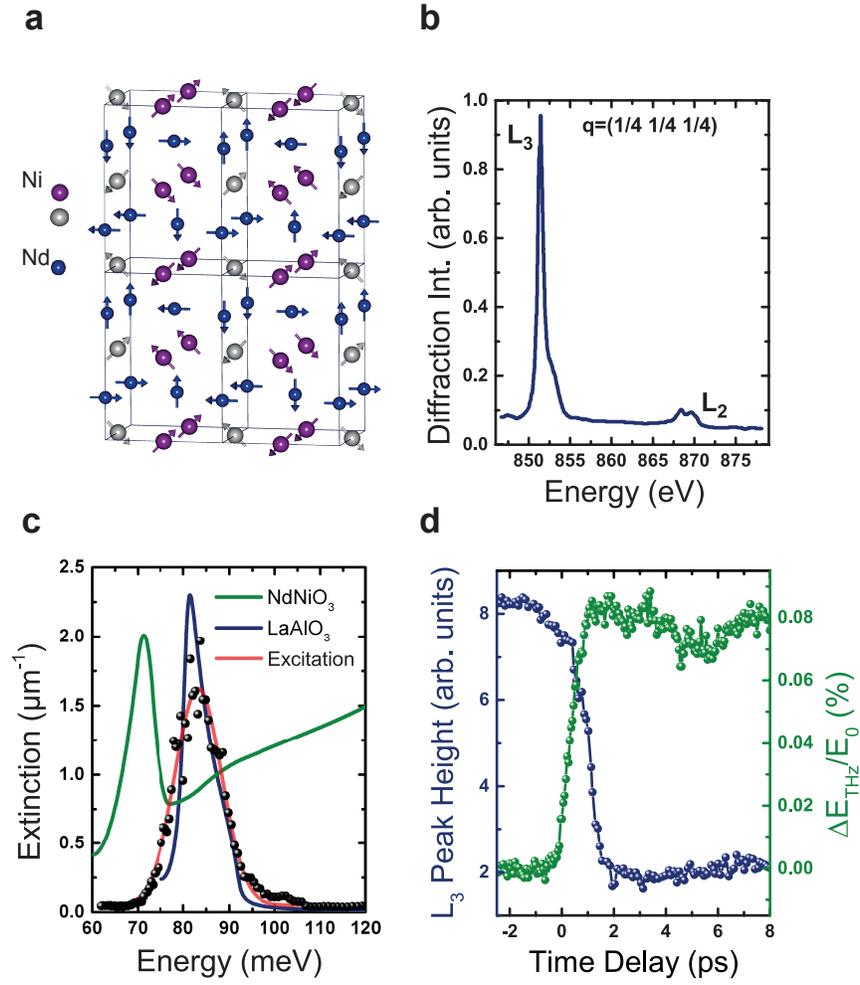


Fig. 2

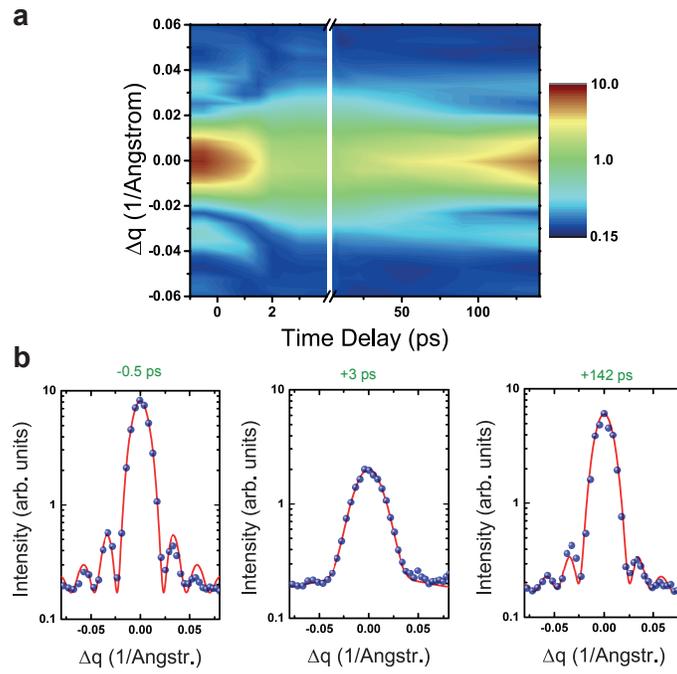


Fig. 3

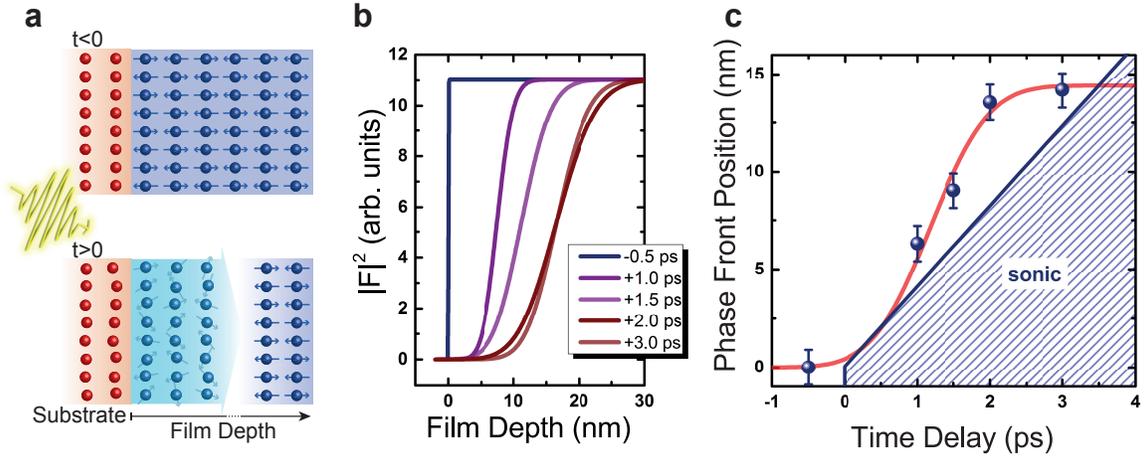


Fig. 4

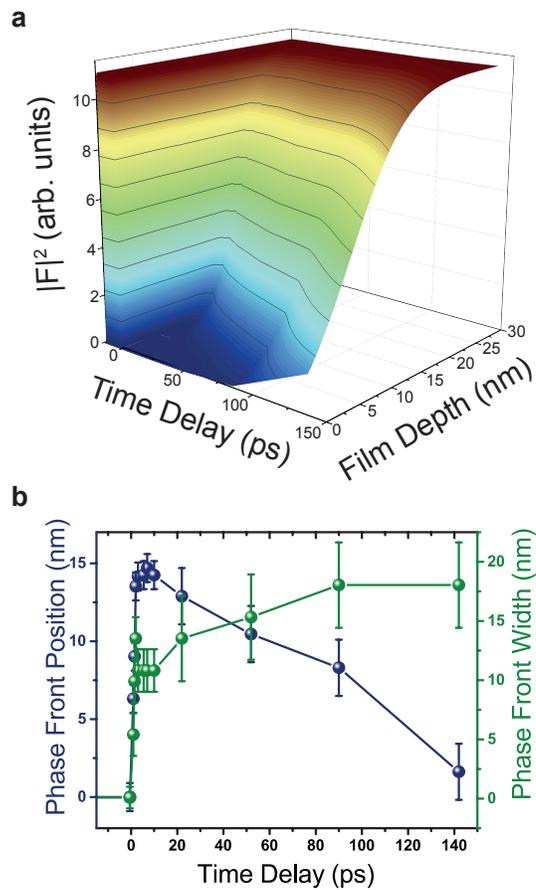
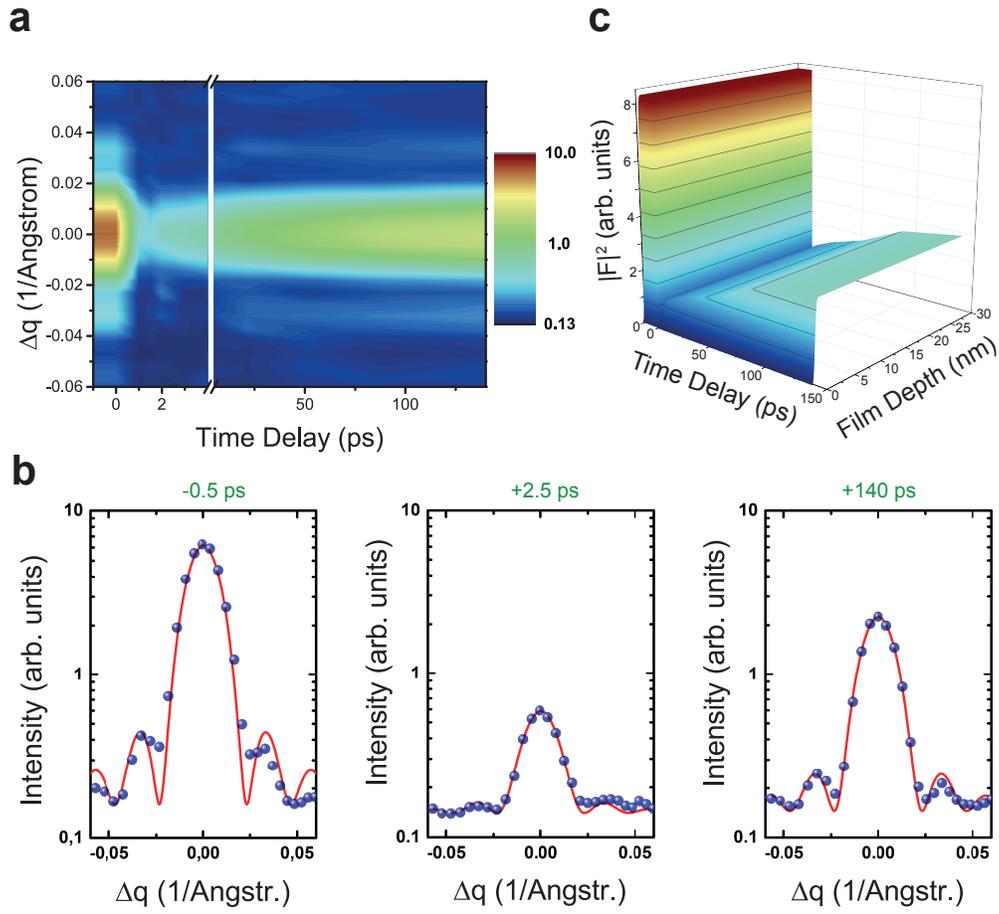


Fig. 5

Near-infrared excitation at 800nm



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