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Bi-directional ultrafast electric-field gating of interlayer charge transport in a cuprate superconductor

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In cuprate superconductors, tunnelling between planes makes three-dimensional superconductive transport possible. However, the interlayer tunnelling amplitude is reduced when an order-parameter-phase gradient between planes is established. As such, interlayer superconductivity along the *c*-axis can be weakened if a strong electric field is applied along the *c*-axis. In this Letter, we use high-field single-cycle terahertz pulses to gate interlayer coupling in $La_{1.84}Sr_{0.16}CuO_4$. We induce ultrafast oscillations between superconducting and resistive states and switch the plasmon response on and off, without reducing the density of Cooper pairs. In-plane superconductivity remains unperturbed, revealing a non-equilibrium state in which the dimensionality of the superconductivity is time-dependent. The gating frequency is determined by the electric field strength. Non-dissipative, bi-directional gating of superconductivity is of interest for device applications in ultrafast nanoelectronics and represents an example of how nonlinear terahertz physics can benefit nanoplasmonics and active metamaterials.

Transport in cuprate superconductors can be understood by considering a stack of intrinsic Josephson junctions made of superconducting planes separated by insulating layers. Three key features characterize the *c*-axis electrodynamics in the superconducting state. First, the d.c. resistivity vanishes, as superconductive tunnelling 'shorts' resistive transport through incoherent quasi-particles. Second, the imaginary part of the conductivity displays a $1/\omega$ frequency dependence, reflecting diamagnetism and the Meissner effect as $\omega \rightarrow 0$. Third, the combination of tunnelling, which has an equivalent inductive impedance, and capacitive coupling between the planes, leads to collective plasma oscillations of superconducting electrons at terahertz frequencies, or Josephson plasma waves[1].

These properties, typically measured in the frequency domain[2], can be observed directly with time-domain terahertz spectroscopy[3]. Figure 1a shows one such electro-optic sampling measurement of a single-cycle terahertz field, after reflection off the optimally doped cuprate $La_{1.84}Sr_{0.16}CuO_4$ (refs 4,5). In the superconducting phase (red curve), long-lived oscillations at a frequency of 2 THz appear on the trailing edge of the pulse. The incident field was measured after reflection from a gold-coated fraction of the sample surface. The frequency-dependent complex reflection coefficient $r(\omega) = E_{refl}(\omega)/E_{inc}(\omega)$, was then derived by dividing the Fourier transforms of the time-dependent reflected field of Fig. 1a by the incident one. The reflectivity $|r(\omega)|^2$ is shown in Fig. 1b, and reproduces well the Josephson plasma edge in this compound.

In Fig. 1c, the complex frequency-dependent dielectric function $\varepsilon(\omega)$ of the equilibrium low-temperature state is presented, which was obtained by fitting the reflectivity $|r(\omega)|^2$ with the two-fluid model[6]. As for any plasmonic response, the real part $\mathrm{Re}\{\varepsilon(\omega)\}$ is negative for $\omega < \omega_p$, where $\omega_p/2\pi = 2$ THz is the frequency of the Josephson plasma resonance in $La_{1.84}Sr_{0.16}CuO_4$. The imaginary part $\mathrm{Im}\{\varepsilon(\omega)\}$ is nearly zero over the entire frequency range, indicating negligible dissipation by non-superconducting quasi-particles. Figure 1d also presents the low-temperature conductivity $\sigma(\omega)$, characterized by a vanishing real part for finite frequencies, and a $1/\omega$ frequency dependence in its imaginary component $\mathrm{Im}\{\sigma(\omega)\} = \rho/4\pi\omega$. Here, ρ is the superfluid density, a measure for the stiffness of the condensate at equilibrium[7]. These equilibrium transport properties have been discussed extensively in the past, especially regarding the controversial role of interlayer tunnelling in high-T_C superconductivity[8–10].

The present work aims at perturbing coherent Josephson coupling without injecting incoherent excitations, to gate *c*-axis transport at high repetition rates. It is known that interlayer transport can be altered statically by the application of magnetic[11] or electric fields[12]. This is possible because tunnelling across a weak link depends on the order-parameter-phase difference between the two superconductors (φ), which is affected by application of external fields[13,14]. To achieve this effect on an ultrafast timescale, interlayer voltage drops of a few to tens of millivolts are needed, corresponding to terahertz-frequency transients with peak electric fields of tens of kV cm^{-1}.

High-field terahertz transients were achieved in our experiments with the tilted-pulse front technique, which was used to generate microjoule single-cycle pulses by optical rectification in $LiNbO_3$ (ref. 15). These pulses were tuned to a centre frequency of 450 GHz, well below the 2 THz Josephson plasma edge (Fig. 2). The gate field wavelength was ~0.65 mm, and could be focused down to spot sizes of ~1 mm^2 to reach field strengths of up to 100 kV cm^{-1}. The gate field was polarized perpendicular to the planes and was completely reflected, penetrating over a distance of 5 μm as an evanescent wave.

The time-dependent reflectivity of $La_{1.84}Sr_{0.16}CuO_4$ was probed both perpendicular and parallel to the planes with a delayed terahertz probe pulse. The probe bandwidth extended up to 2.5 THz and, for *c*-axis polarization, it covered the Josephson plasma edge (Fig. 2). To extract the time-dependent conductivity, the amplitude- and phase-resolved transients were fitted by a model that considered a 5-μm-thick surface layer of unknown conductivity, over an

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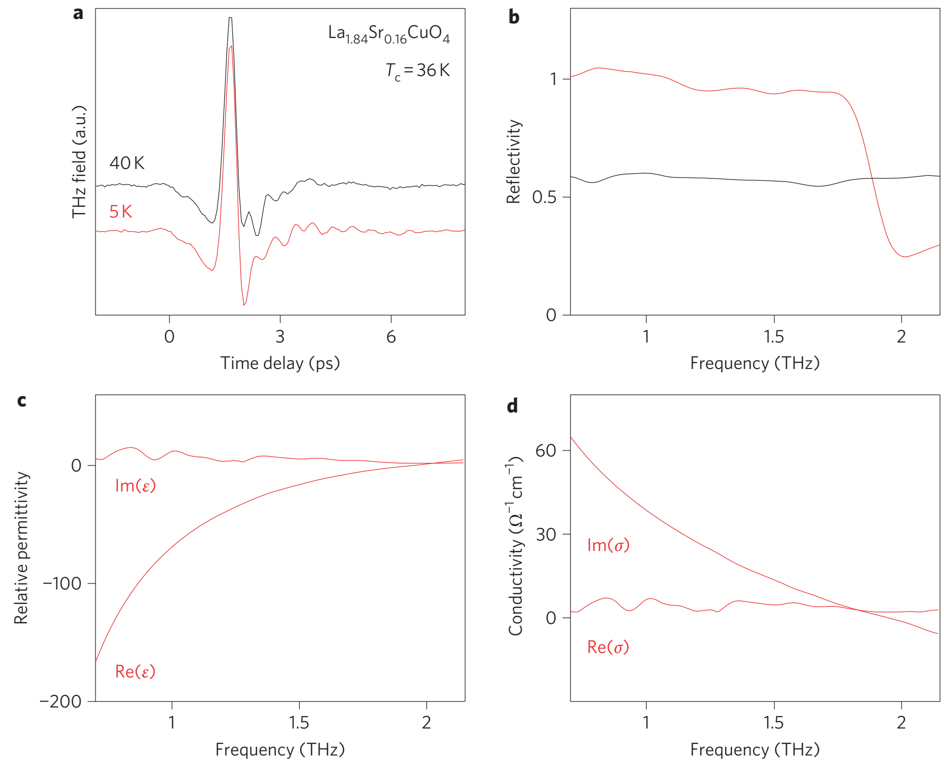

Figure 1 | Equilibrium optical properties of $La_{1.84}Sr_{0.16}CuO_4$. In $La_{2-x}Sr_xCuO_4$, $x = 0.16$ corresponds to the optimal doping level[4]. The anisotropy in this system is a decreasing function of doping, both in the normal and superconducting states. In the normal state, the resistivity ratio decreases for $x > 0.08$, and was measured to be $\rho_c/\rho_{ab} \approx 450$ at $T = 40$ K in the sample used here. In the superconducting state, the anisotropy ratio of the penetration depth is $\lambda_c/\lambda_{ab} \approx 15$ (ref. 5). **a**, Reflected terahertz electric fields from $La_{1.84}Sr_{0.16}CuO_4$, above (black curve, 40 K) and below (red curve, 5 K) $T_C = 36$ K. **b**, Electric field intensity reflectivity spectrum above (black curve, 40 K) and below (red curve, 5 K) $T_C = 36$ K. **c**, Real and imaginary parts of the frequency-dependent equilibrium permittivity $\varepsilon(\omega)$ at 5 K. **d**, Real and imaginary parts of the equilibrium conductivity $\sigma(\omega)$ in the superconducting state at 5 K.

unperturbed semi-infinite superconductor with the optical properties of Fig. 1d.

The key observation of our work is reported in the two-dimensional plots of Fig. 3a, which display the frequency-dependent conductivities (real and imaginary parts) for different time delays τ between a 80 kV cm^{-1} single-cycle gate field and the probe pulse measured in the pump–probe geometry described in ref. 16. In the two upper panels, *c*-axis measurements are displayed. As the gate electric field evolves in time, superconductive coupling vanishes

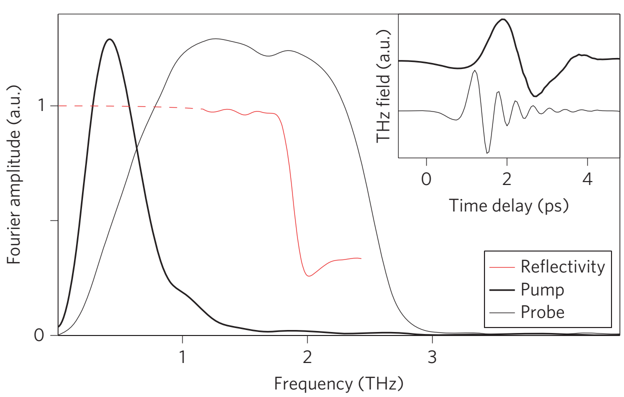

Figure 2 | Characterization of pump and probe pulses. Scaled spectra of terahertz pump (thick black curve) and probe (thin black curve) pulses, overlaid to the $La_{1.84}Sr_{0.16}CuO_4$ reflectivity edge at 5 K (red curve). Inset: typical time-domain transients of terahertz pump and probe pulses measured by electro-optic sampling.

for $\tau = 1.25$ ps, as qualitatively shown by the loss of spectral weight in the imaginary conductivity, and in the corresponding gain of the real part. Remarkably, superconductive transport is re-established within a few hundred femtoseconds ($\tau = 1.5$ ps), when the conductivity of the unperturbed superconductor is recovered. Oscillations between the two states follow. Figure 3b shows lineouts of the complex conductivity at the peaks and troughs of these oscillations. At negative time delays and in the recurring superconducting states, $\mathrm{Re}\{\sigma(\omega,\tau)\}$ nearly vanishes at all frequencies, whereas $\mathrm{Im}\{\sigma(\omega,\tau)\}$ follows a $1/\omega$ frequency dependence, as in Fig. 1d. At time delays where resistive states are established (dashed curve), the real conductivity is the dominant contribution and tends to a finite value σ_0 for $\omega \to 0$, as expected for a Drude gas of incoherent quasi-particles. In this state, the imaginary conductivity still exhibits its $1/\omega$ dependence, but with a strongly depleted pre-factor.

A second important observation results from the *a*–*b*-plane conductivity, which remains essentially unperturbed throughout these dynamics as displayed in the lower panels of Fig. 3a. Effectively, the dimensionality of the superconductivity oscillates in time as the planes are decoupled, an exotic phenomenon never observed to date. The fact that the in-plane optical properties do not show significant change reinforces the notion that the terahertz field perturbs the phase but does not ionize the Cooper pairs, effectively maintaining the modulus of the order parameter unperturbed.

The physics observed here can be quantitatively discussed as follows. The interlayer coupling strength can be described as an equivalent tunnelling inductance L_s, proportional to $1/\cos\varphi$ (ref. 14). At equilibrium $\varphi \approx 0$, the inductance L_s is at a minimum and transport by non-condensed, incoherent quasi-particles is optimally 'shorted'. For weak electric fields, a supercurrent J_s is driven

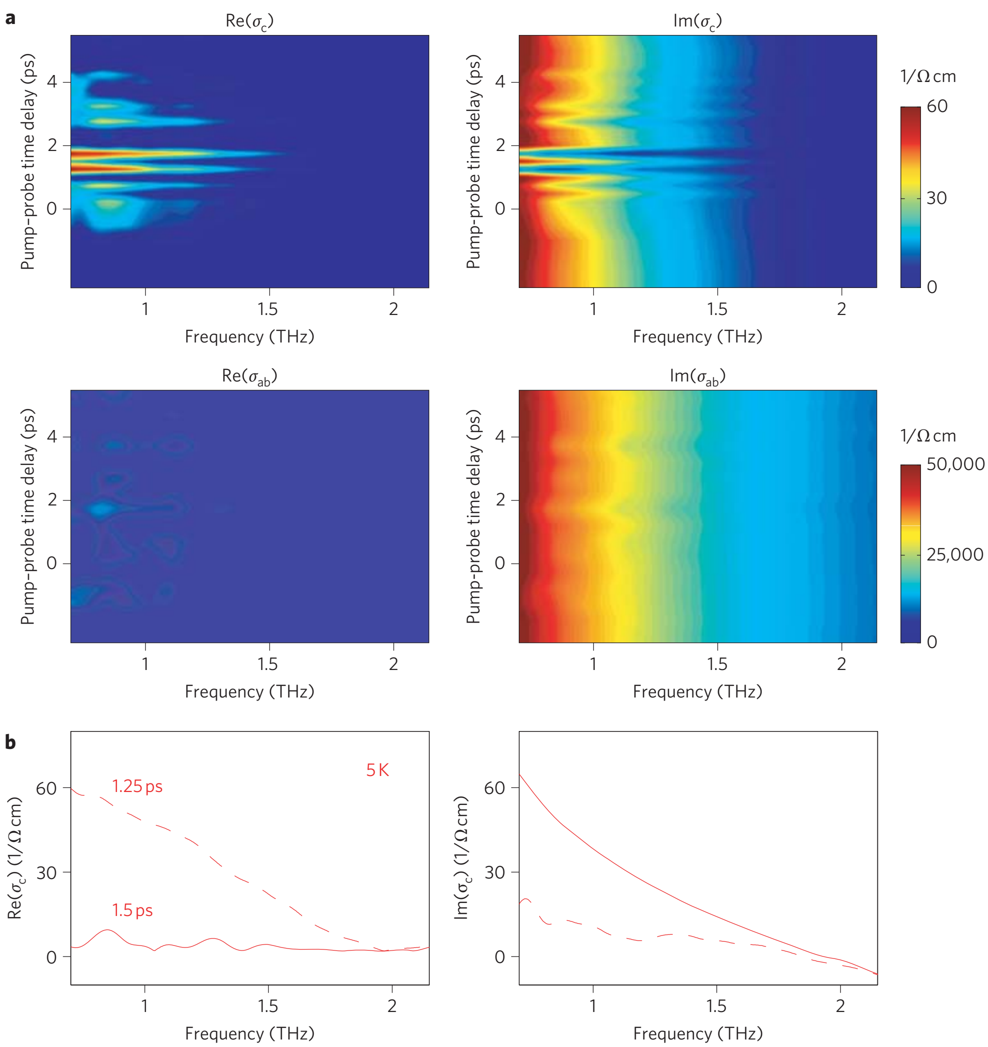

Figure 3 | Time-dependent optical conductivity. a, Upper panels show two-dimensional plots of real and imaginary parts of the *c*-axis conductivity $\sigma_c(\omega,\tau)$ for different pump-probe time delays, measured at 5 K. Pump and probe polarizations are parallel to the *c*-axis. The pump field strength is 80 kV cm^{-1}. For $\tau < 0$, the lineouts of Fig. 3a reflect the unperturbed optical conductivity. As the gate field progresses, the conductivity oscillates rapidly between resistive and superconductive states. Oscillations persist for a couple of picoseconds, comparable to the Josephson coherence time. The lower two panels display the low-temperature in-plane conductivity $\sigma_{ab}(\omega,\tau)$, measured by rotating the probe polarization by 90° to be parallel to the *a*–*b* plane, with the pump polarization kept parallel to the *c*-axis. **b**, Lineouts of conductivity $\sigma_c(\omega,\tau)$ at the peaks and troughs of the oscillations, exhibiting resistive (1.25 ps) and superconductive (1.5 ps) coupling.

through the layers, and a gradient in φ develops across the junction as, according to the first Josephson equation, $J_s \propto \sin\varphi$. For large fields, as L_s increases, ohmic conduction by quasi-particles becomes relevant and a voltage drop V develops across the junction.

At this point, the order parameter phase difference starts advancing in time according to the second Josephson equation, that is, as $\dot{\varphi} = 2eV(t)/\hbar$, where e is the electron charge and $\hbar$ Planck's constant. The tunnelling then evolves as $L_s \propto 1/\cos\varphi$, diverging when φ crosses $\pm\pi/2$, and changing sign in between.

In the upper panel in Fig. 4a, we report the normalized time integral of the gate electric field $\int E_{\mathrm{pump}}(t)\mathrm{d}t$, derived from the measured terahertz pump transients $E_{\mathrm{pump}}(t)$. The integral of the field is compared to the measured time-dependent strength of *c*-axis superconducting transport, evaluated as $S(\tau) = \lim_{\omega\to 0}\omega\mathrm{Im}(\sigma(\omega,\tau))$. At equilibrium, this quantity is proportional to the superfluid density ρ. Here, $S(\tau)$ is used as a measure of the non-equilibrium interlayer coupling strength and is well-fitted by the function $|\cos(c\int E_{\mathrm{pump}}(t)\mathrm{d}t)|$ with only the constant c left as a free parameter. As the integral of the driving voltage evolves, and as φ crosses $\pi/2$, $S(\tau)$ vanishes, recovering as φ is driven towards π. The model does not fit well at low field strengths, because, according to the discussion above, the transport is still superconducting and no voltage drop develops. At high fields, the model faithfully reproduces the measured non-equilibrium interlayer coupling strength $S(\tau)$ over a large dynamic range.

The superconductive–resistive oscillation frequency, measured by sampling the peak of the probe terahertz transient as a function of delay, is shown in Fig. 4b as a function of pump field strength. Two regions are identified. Below $E^* \approx 75$ kV cm^{-1}, the transport is modulated at the frequency of the pump. In this regime the interlayer phase difference is perturbed but does not reach $\pi/2$, and the coupling is never completely shut off. For field strengths above E^*, the modulation frequency increases linearly with the field, in the spirit of the a.c. Josephson effect.

This voltage-to-frequency conversion is a new demonstration of nonlinear terahertz physics that could be extended to photonic devices and modulators, and also to nanoplasmonic devices. We also note its potential applications to electronics. Because interlayer

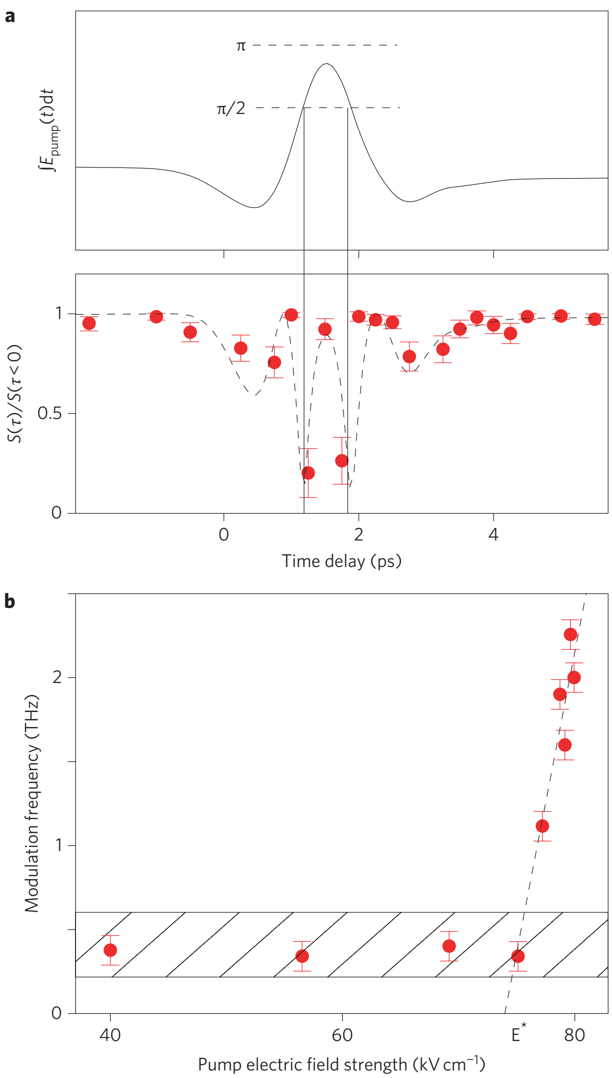

Figure 4 | Ultrafast electric field gating. a, The upper panel shows the normalized integral of the measured terahertz pump transient as a function of time delay, which is proportional to the advancement of the interlayer phase difference $\varphi(t) \propto \int^{t} E_{pump}(t')dt'$. The lower panel shows $S(\tau) = \lim_{\omega \to 0} \omega \mathrm{Im}(\sigma(\omega, \tau))$ as a measure of interlayer coupling strength extracted from our experimental data (red dots), together with error bars obtained from the low-frequency extrapolation. The data points are fit by $|\cos(c \int E_{pump}(t)dt)|$ (black dashed line), where c is left as a free parameter. **b**, Modulation frequency dependence on the terahertz gate electric field strength with estimated systematic error. For low amplitudes $E_{pump} < E^*$, the modulation occurs at the pump frequency (the shaded area represents the full-width at half-maximum of the probe amplitude spectra). Above the critical threshold E^*, the modulation frequency increases linearly with field strength, reminiscent of the a.c. Josephson effect.

transport is determined by short-range tunnelling between neighbouring layers, this effect could be applied to single nanoscale junctions. Inspired by the present work, it would be interesting to explore terahertz pulse sequences with voltage integrals that drive the phase by multiples of $\pi/2$ to switch the dimensionality at will. These experiments would be relevant below and above the Berezinskii–Kosterlitz–Thouless temperature, corresponding to different regimes of stability for two-dimensional superconductivity. Finally, strong field perturbations of interlayer couplings[17] may be used to test new ideas of the physics of cuprates, including the case of striped states for which Josephson de-coupling may be important[18].

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Author contributions

A.C. conceived the project. A.D. designed and executed the experiment and analysed the data. A.D. and A.C. interpreted the data. M.C.H., D.F. and J.C.P. assisted in the experimental realization. S.P., T.T. and H.T. grew the samples.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper at www.nature.com/naturephotonics. Reprints and permission information is available online at http://www.nature.com/reprints/. Correspondence and requests for materials should be addressed to A.D. and A.C.