Supplementary material for "Diffusion of Nonequilibrium Quasiparticles in a Cuprate Superconductor"

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1 Experimental method

In the following we describe the procedure to measure the index of refration grating created by the pump beams. We first block the reflected beam labelled P2 in Fig. 1. In this configuration, the electric field at the detector is the sum of the reflected P1 and the diffracted P2,

$$E_D = E_{P1}(r_0 + \delta) + E_{P2}\eta \tag{1}$$

where r_0 is the equilibrium reflection coefficient, δ is the q=0 component of the change in reflection coefficient, and η is the $q = 2\pi/\lambda_g$ component. The output current of the Si detector, I_D , is proportional to $|E_D|^2$,

$$I_D \propto |E_{P1}r_0|^2 + 2|E_{P1}r_0|^2[|\delta|\cos(\phi) + |\eta|\cos(\phi - \theta)],$$
(2)

where ϕ is the phase of δ relative to that of r_0 , while θ is the phase of E_{P1} relative to E_{P2} . In obtaining Eq. 2, terms that are quadratic in δ or η are neglected as these coefficients are of order 10^{-5} or less, and we set $|E_{P1}| = |E_{P2}|$ since the two probe beams have equal intensity. If the other return path is blocked instead, the equation for the current is the same except that the sign of θ is reversed

$$I_D \propto |E_{P1}r_0|^2 + 2|E_{P1}r_0|^2[|\delta|\cos(\phi) + |\eta|\cos(\phi + \theta)]$$
(3)

In Eq. 3 the first term on the right side is the reflected intensity in equilibrium and the second term is the change in reflectivity that we wish to measure. To isolate the terms proportional to δ and η we modulate the amplitude of the pump at 100 kHz. Lock-in detection at the modulation



Figure 1: Illustration of the apparatus for heterodyne transient grating detection. Pump and probe beams from the laser are split at the diffractive optic (for clarity only the probe beams are shown). A spherical mirror and plane folding mirror (represented schematically by a lens in the sketch) focus the beams to a single 100 μ m spot on the sample. After specular reflection and diffraction at the sample surface, the two probe beams are recombined by the diffractive optic and directed to a Si photodiode detector. The wavevector of the quasiparticle density variation is changed, without optical realignment, by translating the diffractive optic so that a different phase mask in the array is inserted in the beam.

frequency removes the first term in I_D , which does not depend on the pump intensity. The current that is proportional to $|E_{P1}r_0|^2$ is determined from the nonmodulated portion of the detector current. Dividing the modulated component by the nonmodulated component yields $|\delta| \cos(\phi) + |\eta| \cos(\phi \pm \theta)$.

The next step is to determine θ , the relative phase of the two probe beams. The procedure is to measure the lock-in output as a function of θ . To vary θ a coverslip is placed in each of the two probe beam paths (see Fig. 1). One coverslip is fixed and the other is mounted on a rotational stage. Fig. 2 shows the dependence of the lock-in output on the angle of the rotating coverslip. Each curve in the figure is the lock-in output as a function of time-delay between pump and probe pulses, for a given coverslip angle. To acquire the sequence of transients the coverslip was rotated in steps of 0.07° which corresponds to a phase advance of approximately 0.3 radians.



Angle (θ)

Figure 2: Calibration of the phase. Fractional change in reflectivity, $\Delta R/R_0$, of the two different probes P1 (top panel) and P2 (bottom panel) as a function of time is plotted at different cover slip angles.

We fit the peak values of the transients to the form expected for the two paths, that is $|\delta|cos(\phi) + |\eta|cos(\theta \pm \phi)$. The fit determines a calibration between coverslip angle and θ . Using this calibration we establish the angles that correspond to $\theta = 0, \pi/2$ and π . Performing measurements at these three values of θ , we obtain $|\delta|cos(\phi) \pm |\eta|cos(\phi)$ and $|\delta|cos(\phi) + |\eta|sin(\phi)$, from which the three parameters that characterize the grating, $|\delta|, |\eta|$, and ϕ are readily determined. In the paper we refer to $|\delta|$ as R, and $|\eta|$ as TG.

2 Modeling

In the main text we refer to theoretical modeling of the evolution in space and time of the nonequilibrium quasiparticle density. The modeling was performed by numerically integrating the equation,



Figure 3: Evolution of the quasiparticle concentration (light-blue shading) in the recombination-dominated regime according to the equation given in the text.

$$\partial n(x,t)/\partial t = D\partial^2 n(x,t)/\partial x^2 - \beta n^2(x,t), \tag{4}$$

where n is the quasiparticle density, D is the diffusion coefficient, and β is the recombination coefficient. Fig. 3 shows results for initial conditions that correspond to the high density regime, $\beta n \gg Dq^2$. The light blue shading corresponds to the quasiparticle density. The pattern on the top is the initial density profile and the patterns below show its evolution in time. After creation, recombination is most rapid in the regions of highest quasiparticle density. The density-dependent recombination distorts the sinusoidal grating by flattening the crests. This leads to decay of TG/Reven in the absence of diffusion. The grey regions show the energy accumulated as the quasiparticles recombine, which is proportional to n(x,0) - n(x,t). The recovery of TG/R to its initial value at long times indicates that this energy is stored in non-propagating modes which also induce a change in the index of refraction. When all of the energy originally stored in the quasiparticles transfers to such modes, the index grating recovers its initial sinusoidal form, and TG/R approaches its value at time zero.