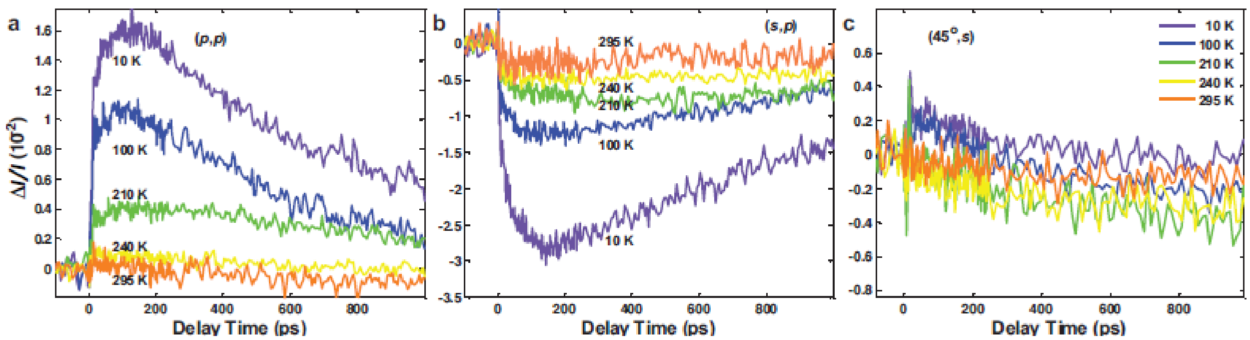

Supplementary Figure 1. The evolution of the photoinduced changes in SHG intensity ($\Delta I/I$) versus delay time and temperature, mediated through strain. $\Delta I/I$ from polar combinations of (a) (*p*,*p*), (c) (*s*,*p*), and (c) (45^o,*s*) are shown, measured at various temperatures.

Supplementary Table 1.
The various polarization combinations probe the coefficients (a, b, c), allowing us to extract (d_{15}, d_{31}, d_{33}). Each coefficient (a, b, c) is a function, f, of (d_{15}, d_{31}, d_{33}) and θ.

	(45^{o}, s)	(s, p)	(p, p)
SHG dipole	$P_y(2\omega)$	$P_z(2\omega)$	$P_x(2\omega)$, $P_z(2\omega)$
(a, b, c)	c=f(d15) > 0	b=f(d31) < 0	a=f(d15, d31, d33) > 0
(d_{15}, d_{31}, d_{33})	d_{15} =-1.2	d_{31} =-1	d_{15} =-1.2, d_{31} =-1, d_{33} = 7

Supplementary Note 1. General picture of quasiparticle relaxation in ferromagnetic manganites

Quasiparticle relaxation in a FM manganite (e.g., LCMO) after photoexcitation is governed by the following major processes, roughly separated in time: (a) electron-electron (*e-e*) scattering (<100 fs), (b) electronphonon (*e-ph*) coupling (~1 ps), (c) spin-lattice (*s-l*) relaxation (~30-100 ps), and (d) thermal diffusion (>0.5 ns), both within the film and in the substrate. Initially, the photoexcited quasiparticles have ~1.5 eV per electron-hole pair. After *e-e* scattering, the quasiparticles develop a Fermi-Dirac distribution with a defined electronic temperature T_e. This process is very fast (within a hundred femtoseconds) and is usually not resolved within the laser pulse width (~100 fs). The excited quasiparticles can then interact with the spins and lattice. For a quasiparticle to flip its spin, the total angular momentum is conserved, which in turn only allows demagnetization through exchange of angular momentum between the spin and orbital degrees of freedom [1, 2]. Therefore, the energetic carriers first relax through *e-ph* coupling, which increases the lattice temperature, t ~1 ps in Fig. 2a. Now the lattice can interact with the spins, and through phonon or impurity scattering the spin-flip process is allowed for spin relaxation, occurring on a longer timescale. This process therefore demagnetizes spins within tens of picoseconds [3-5], t~50 ps in Fig. 2a. LCMO then reaches quasi-equilibrium through a thermal gradient that slowly diffuses towards both the substrate and BSTO layer, finally dissipating through the substrate (t >0.5 ns in Fig. 2a).

We performed optical pump-probe measurements (with 1.59 eV pump and 3.18 eV probe energies) to directly measure quasiparticle relaxation in our BSTO/LCMO heterostructure, which can provide additional insight through comparison with our time-resolved SHG data (Fig. 4). After 1 ps, the photoinduced change in SHG intensity, $\Delta I/I$, for (incident, detected) (p, p) and (s, p) polarizations, as well as the photoinduced change in reflectivity, $\Delta R/R$, at low temperatures can be fit using two exponential processes with time constants of $\tau_{s\text{-}l}$ ~30-45 ps and τ_D ~450-650 ps. The fast sub-ps relaxation in $\Delta R/R$, due to *e-ph* coupling, is not observed in $\Delta I/I$, as expected from the discussion in the main manuscript. The room temperature $\Delta I/I$ data is best reproduced with a rise time ~7 ps and a decay time ~600 ps (which approximately fits all polar combinations), while the $\Delta R/R$ data is best fitted with a rise time ~1 ps and a decay time ~500 ps.

Supplementary Note 2. Magnetostriction

The simplest Hamiltonian associated with magnetostriction from t_{2g} spins and the lattice is

$$H = \Sigma_{ij} \tfrac{1}{2} k r_{ij}^2 + \Sigma_{ij} J_{ij}(\mathbf{r}_{ij})\mathbf{S}_i\mathbf{S}_j, \quad (1)$$

where i, j are different atoms. When the spins are in an ordered state, the spin-spin interaction $J_{ij}(\mathbf{r}_{ij})$, irrespective of parallel ($\mathbf{S}_i\mathbf{S}_j>0$, $J_{ij}(\mathbf{r}_{ij})<0$) or antiparallel ($\mathbf{S}_i\mathbf{S}_j<0$, $J_{ij}(\mathbf{r}_{ij})>0$) alignment, energetically favors lattice contraction. This is due to the overall negative sign of the second term in Eq. (1). In addition, lattice contraction increases $J_{ij}(\mathbf{r}_{ij})$, lowering the total energy. In thermodynamic equilibrium at T_C, the second term of Eq. (1) induces a non-analytic point in the lattice constant and the correlated spins contract the lattice upon cooling [6].

Supplementary References

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