Citation

Mazzone, Daniel G.; Meyers, Derek; Cao, Yue; Vale, James G.; Dashwood, Cameron D.; Shi, Youguo; James, Andrew J. A.; Robinson, Neil J.; Lin, Jiaqi; Thampy, Vivek; Tanaka, Yoshikazu; Johnson, Allan S.; Miao, Hu; Wang, Ruitang; Assefa, Tadesse A.; Kim, Jungho; Casa, Diego; Mankowsky, Roman; Zhu, Diling; Alonso-Mori, Roberto; Song, Sanghoon; Yavas, Hasan; Katayama, Tetsuo; Yabashi, Makina; Kubota, Yuya; Owada, Shigeki; Liu, Jian; Yang, Junji; Konik, Robert M.; Robinson, Ian K.; Hill, John P.; McMorrow, Desmond F.; Först, Michael; Wall, Simon; Liu, Xuerong and Dean, Mark P. M. (2021). Laser-induced transient magnons in Sr3Ir2O7 throughout the Brillouin zone. Proceedings of the National Academy of Sciences of the United States of America, 118(22), article no. e2103696118.

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Laser-Induced Transient Magnons in Sr$_3$Ir$_2$O$_7$
Throughout the Brillouin Zone

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This manuscript was compiled on May 14, 2021

Although ultrafast manipulation of magnetism holds great promise for new physical phenomena and applications, targeting specific states is held back by our limited understanding of how magnetic correlations evolve on ultrafast timescales. Using ultrafast resonant inelastic x-ray scattering we demonstrate that femtosecond laser pulses can excite transient magnons at large wavevectors in gapped antiferromagnets, and that they persist for several picoseconds which is opposite to what is observed in nearly gapless magnons. We find that transient magnetic fluctuations are trapped throughout the entire Brillouin zone, while remaining present beyond the time that is required to restore the original spin network. The results are interpreted in the context of a spin-bottleneck effect, in which the existence of an explicit magnetic decay channel allows for an efficient thermalization of transient spin waves.

Significance Statement
Ultrafast manipulation of magnetic states holds great promise for progress in our understanding of new quantum states and technical applications, but our current knowledge of transient magnetism is very limited. Our work elucidates the nature of transient magnetism in gapped antiferromagnets using Sr$_3$Ir$_2$O$_7$ as a model material. We find that transient magnetic fluctuations are trapped throughout the entire Brillouin zone, while remaining present beyond the time that is required to restore the original spin network. The results are interpreted in the context of a spin-bottleneck effect, in which the existence of an explicit magnetic decay channel allows for an efficient thermalization of transient spin waves.

www.pnas.org/cgi/doi/10.1073/pnas.XXXXXXXXXX
PNAS | May 14, 2021 | vol. XXX | no. XX | 1–6


The authors declare no competing interests.

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Demagnetization pathways in antiferromagnets. (A) Multiple spin configurations can give rise to the same macroscopic magnetization. These are indistinguishable in order parameter measurements commonly probed in ultrafast studies. Three such cases are sketched, corresponding to a reduced magnetic moment, a collective spin rotation and a disordered state. The first two cases are uniform perturbations to the entire spin network, whereas the latter is due to short range disorder of individual spins. (B) Relative magnetic (3.5, 1.5, 18) Bragg peak intensity in Sr$_2$IrO$_7$ as a function of time delay [notation in reciprocal lattice units (r.l.u)]. The data are plotted up to 7 ps after the arrival of the optical pump at $t=0$. The error bars follow Poissonian statistics.

The energy loss and momentum change of scattered photons enabling to probe short-range magnetic fluctuations, such as magnons, in the transient state. Tr-RIXS measurements of magnons are extremely challenging technically. The first experiment was performed a few years previously on Sr$_2$IrO$_4$, but the only observable effects occurred at the magnetic ordering wavevector (2). This appears superficially congruent with the idea that the effectively zero momentum light can only excite individual magnons at the center of the magnetic Brillouin zone. Here we study gapped antiferromagnet Sr$_2$Ir$_2$O$_7$, and directly show that photo-excitation can modify magnons throughout the Brillouin zone which persist beyond a picosecond timescale. We suggest that the large spin gap in Sr$_2$Ir$_2$O$_7$ blocks the cooling of transient magnons that appears to occur in Sr$_2$IrO$_4$.

Demagnetization pathways in antiferromagnets

Figure 1B shows the time evolution of the Sr$_2$Ir$_2$O$_7$ magnetic Bragg peak intensity up to 7 ps after the arrival of the 2 µm (620 meV) laser pump that excites carriers across the band-gap (18). This was measured using Ir $L_{3}$-edge tr-REXS at $T \approx 110$ K $\ll T_N = 285$ K using the (-3.5, 1.5, 18) magnetic Bragg peak (see Materials and Methods). Immediately after the pump, magnetism is reduced by 50-75% for laser fluences between 5.2 and 32.2 mJ/cm$^2$. This occurs faster than our time resolution of 0.15 ps, which is substantially quicker than in other materials including Sr$_2$IrO$_4$ (19). It is noted that even for strong fluences some remnant magnetic fraction is observed immediately after the laser pump, suggesting some experimental mismatch between pumped laser and probed x-ray volumes (c.f. SI Appendix). However, the transient signal shows signs of saturation, suggesting the pumped volume is completely demagnetized. We refer to further details on the recovery of the magnetic long-range order in the SI Appendix. The tr-REXS results in Fig. 1B clearly show that a 2 µm laser pump results in an appreciable quenching of magnetic long-range order that survives on a picosecond timescale. However, the experiment does not reveal the microscopic spin configuration of the transient state. As illustrated in Fig. 1A a transiently suppressed magnetic order parameter can arise from several very different microscopic configurations that give the same REXS response. We thus studied the RIXS response of the transient state to gain insight into the Q dependence of the magnetic correlations.

Transient evolution of magnetic short-range correlations

Figures 2A and B display energy-loss spectra of Sr$_2$Ir$_2$O$_7$ measured at $(\pi, \pi)$ and $(\pi, 0)$, corresponding to the magnetic Brillouin zone center and zone boundary. The blue data points correspond to RIXS spectra in the equilibrium state, showing four features; an elastic line, a collective magnon, a magnon continuum and an orbital excitation (20–23). The collective magnon features a moderate dispersion, shifting in energy from 100 meV at $(\pi, \pi)$ to 150 meV at $(\pi, 0)$, while the magnon continuum (250 meV) and orbital excitation (680 meV) reveal negligible dispersion. It is believed that the magnetic excitations in equilibrium arise predominantly from the combination of Heisenberg-like interactions within the iridium layers and an anisotropic exchange perpendicular to the tetragonal plane (see also SI Appendix) (20, 24, 25). The orbital excitation, on the other hand, can be understood considering the electronic configuration of Sr$_2$Ir$_2$O$_7$. It is defined by the five iridium valence electrons, residing in a crystal-field-derived $t_{2g}$ orbital manifold. The ground state is further split by strong spin-orbit coupling and moderate Coulomb interactions, establishing a filled $J_{\text{eff}} = 3/2$ and half-filled $J_{\text{eff}} = 1/2$ Mott state (20, 24–26). Thus, the excitation epitomize a transition between these two manifolds (20, 22, 23).

We have overplotted the energy-loss spectra in equilibrium (blue) with transient state data (orange). These have been prepared with a 20 mJ/cm$^2$ laser pulse, for which a substantial suppression of magnetic order has been found (see Fig. 2B). Because small drifts in the X-ray source position can result into shifts of the measured RIXS spectrum, static and pumped spectra were measured with alternate shots of the free electron laser for each delay. The time resolution of the setup equaled ~400 fs (see Materials and Methods for details). At both reciprocal lattice positions, we observe significant changes in the electronic correlations at $t > 0$. This is seen most clearly in the difference spectra shown in Figs. 2C and D, evincing that the magnon and orbital excitations are altered upon photo-doping at both, $(\pi, \pi)$ and $(\pi, 0)$.

Further insight into the transient short-range correlations
Fig. 2. Static and transient electronic short-range correlations of Sr$_3$Ir$_2$O$_7$. Tr-RIXS spectra at the minimum (A) and maximum of the dispersion (B). The spectra show an elastic line, a magnon, a magnon continuum and an orbital excitation at $\sim$680 meV, before the arrival of the laser pump (blue) and for different time delays (orange). Errorbars are determined via Poissonian statistics. The dotted lines are contributions of best fits to the data (solid lines), consisting of four Gaussians and a constant background. Their difference spectra are shown in (C) and (D).

Discussion

The cross section of the orbital excitation around 680 meV is dependent on the respective charge occupations of the $J_{\text{eff}} = 3/2$ and $J_{\text{eff}} = 1/2$ state. The intensity would, for instance, decrease if fewer than four electrons per Ir side reside in the $J_{\text{eff}} = 3/2$ ground state, and increase if the $J_{\text{eff}} = 1/2$ state was empty. The transient depletion of the excitation thus provides direct evidence that not only $J_{\text{eff}} = 1/2$ but also $J_{\text{eff}} = 3/2$ electrons are pumped over the insulating band gap, and that some of the $J_{\text{eff}} = 3/2$ carriers do not decay into their orbital ground state within 5 ps, which is in line with the incomplete recovery of magnetic long-range order (see Fig. 1B).

The substantial anisotropy perpendicular to the tetragonal plane leads to collinear magnetic long-range order, revealing a large magnon gap that is minimal at $(\pi, \pi)$ and maximal at $(\pi, 0)$ (cf. Fig. 4B for details) (20–24). Thus, the results in Fig. 3A show that magnons are modified over an extended area in reciprocal space, and that these changes persist for several picoseconds. This experimental result is strikingly different from the behavior in materials hosting a gapless excitation spectrum. In isotropic Heisenberg-like Sr$_2$IrO$_4$ (see Fig. 4A), for instance, the only observable changes in the magnon occurred at the magnetic Brillouin zone center of $(\pi, \pi)$ (2). In the following discussion, we aim to assess these differences.

The transient state created here features suppressed magnetic long- and short-range correlations throughout the Brillouin zone and it, therefore, has some similarities to a thermal state at elevated temperature. This state, however, appears to form effectively instantaneously, within our time resolution of either 150 or 400 fs depending on whether one considers...
the REXS or RIXS signatures. These timescales are much faster than typical thermalization processes in magnetic insulators, ranging from several to hundreds of picoseconds (27–30). We thus consider the physical mechanisms at play, beyond a simple effective temperature model.

The photo-excitation process promoting charge carriers across the insulating gap leaves behind local spin vacancies that create non-thermal magnons spread across the entire Brillouin zone. These high-energy magnons can decay into lower-energy multiple-particle excitations via magnon-magnon scattering processes following the dispersion relation - which, in lowest approximation, is similar to the one of the unperturbed state. A full thermalization of the system, however, requires a coupling other subsystems, which becomes unavoidable for the dissipation of transient magnons around the minimum of the dispersion relation.

In gapless antiferromagnets, such as Sr₂IrO₄ (25, 31), a steep magnon dispersion from high to effectively zero energy establishes a well-defined decay channel (see Fig. 4a). Thus, spin waves at high energy face multiple possibilities to disintegrate, but the options become increasingly limited around the minimum of the Goldstone-like mode. This gives a natural explanation of why ultrafast magnons were observed only at (π, π), but not at (π, 0) for Sr₂IrO₄ (2). Furthermore, the small spin-wave gap enables an efficient energy transfer into the lattice subsystem via low-energy phonons, restoring the equilibrium configuration.

In summary, we report an instantaneous creation of transient magnons in the gapped antiferromagnet Sr₃Ir₂O₇ after a laser excitation of 2 μm. The ultrafast transverse short-range correlations occur at both extremes of the magnetic dispersion in momentum space, showing the existence of trapped spin waves that are likely present throughout the entire Brillouin zone. This demonstrates an incoherent response of transient magnetism, which is fundamentally different than in gapless antiferromagnets such as in Heisenberg-like Sr₂IrO₄ (2). The

We note that Néel order in Sr₂IrO₄ is unstable if one considers only a single Ir-layer, but stabilized by a very small inter-layer c-axis coupling amplified by a divergence in the in-plane magnetic correlation length within the Ir-layers (see SI Appendix). This is in strong contrast to bi-layer materials such as Sr₃Ir₂O₇ which order magnetically without necessarily invoking coupling between different bi-layers. Although the bi-layer system features far stronger anisotropy and intra-bilayer coupling, this is offset by the spin gap impeding the growth of the in-plane magnetic correlation proceeding the transition. Empirically, Sr₃Ir₂O₇ and Sr₂IrO₄ feature comparable Néel temperatures (285 and 240 K, respectively) despite the spin-wave gap of the former material being roughly 150 times larger (see Fig. 4) (20, 25, 31). Thus, no pronounced decay channel is present in Sr₃Ir₂O₇, which inherently limits the decay of transient magnons along a particular wavevector. Furthermore, phonon-assisted energy transfer processes are strongly reduced by the large spin gap, leading to long-lived magnons that are trapped over a large reciprocal space area.

Following this argument, the partial recovery of magnetic long-range order at a picosecond timescale may arise from two mechanisms. The magnetic time constant is close to the charge timescale that has been reported previously via ultrafast reflectivity measurements (32). This may attest a link between charge and magnetic degrees of freedom, which is also supported by the wavevector-independent suppression of the orbital excitation below 2 ps (see Fig. 3B). In Mott materials the (pseudo-)spin configuration is directly related to the charge distribution on the atomic sites, whose recovery reinstates the original one spin/site population. Thus, it is conceivable to attribute the partial recovery of magnetic order in Sr₃Ir₂O₇ to its Mott nature. Alternatively, the partial recovery of magnetic long-range order may originate from a redistribution of transient magnons within the Brillouin zone, pointing towards a scenario in which the laser pump generates an excess of magnons at the magnetic wavevector. In fact, the results shown in Fig. 3A suggest a suppression of spin waves at (π, π), which is absent at (π, 0).

In summary, we report a near instantaneous creation of transient magnons in the gapped antiferromagnet Sr₃Ir₂O₇ after a laser excitation of 2 μm. The ultrafast transverse short-range correlations occur at both extremes of the magnetic dispersion in momentum space, showing the existence of trapped spin waves that are likely present throughout the entire Brillouin zone. This demonstrates an incoherent response of transient magnetism, which is fundamentally different than in gapless antiferromagnets such as in Heisenberg-like Sr₂IrO₄ (2).
results are interpreted in the context of a spin-bottleneck effect. In Sr₂IrO₄, a steep dispersion exists from high energy magnons at the magnetic zone boundary into low-energy magnons at (π, π), which allows for a highly efficient magnon decay. In contrast, the large magnon gap and moderate dispersion of Sr₃Ir₂O₇ leads to transient spin waves that are trapped in the entire Brillouin zone. Thus, our results emphasize the need to include contributions like anisotropic magnetic exchange interactions alongside electron-phonon couplings in theoretical approaches that model transient magnetic states. Finally, we remind the reader that neither a collinear antiferromagnetic spin alignment, nor magnons are exact eigenstates of a $S = 1/2$ quantum magnet. Thus, it may be natural to expect strong magnon-magnon scattering occurs in some transient states. Further experimental and theoretical efforts along these lines will be vital to progress in the understanding of the ultrafast dynamics in quantum materials, including cases where complex competions of macroscopic quantum phases appear via photo-doping.

Materials and Methods

Sample preparation and characterization. All measurements were performed on Sr₂IrO₄ single crystals that were grown via a flux method as described in Ref. (33), and references therein. The high quality of the crystals were confirmed via Laue and single crystal diffraction. Here, we denote reciprocal space using a pseudo-tetragonal unit cell with $a = 3.896$ Å and $c = 20.88$ Å at room temperature.

All experiments were conducted on the same sample with crystal orientations [1,0,0] and [0,0,1], or [1,1,0] and [0,0,1] in the scattering plane for tr-REXS or tr-RIXS experiments, respectively. In both cases, a horizontal scattering geometry was used and the sample was cooled to $T \approx 110$ K via nitrogen cryostreams.

Optical laser pump. 100 fs long pump pulses of 620 meV (2 μm) were generated by an optical parametric amplifier similar to previous ultrafast experiments on Sr₂IrO₄ (2). The angle between the incident x-ray and infrared pulses was 20°. This leads to an estimated laser penetration depth of ~82 nm for grazing incident x-rays (see SI Appendix).

Time-resolved resonant elastic scattering (tr-REXS). The ultrafast recovery of magnetic long-range order was studied at the pump-probe station from Pring-8 Angstrom Compact free-electron LASer (SACLA) featuring a repetition rate of 30 Hz (34, 35). The x-ray energy was tuned to the Ir $L_{3\text{-}}$edge at 11.215 keV, and refined via a resonant energy scan of the (~3.5, 1.5, 18) magnetic Bragg peak. The reflection was detected at $2\theta = 92.2°$ using a multi-port charged coupled device (MPCCD) area detector (0.0086 r.l.u/pixel) (36). The Bragg condition was established by rotating the sample surface normal $\Delta \chi = 21.2°$ out of the scattering plane. The x-rays were focused to a spot size of 20 μm full-width at half-maximum (FWHM) and hit the sample at a grazing angle of ~1.6°. The x-ray penetration depth was estimated to 162 nm (see SI Appendix). The laser spot size was ~230 μm FWHM at normal incidence. The detector was read out shot-by-shot and thresholded to reduce background arising from x-ray fluorescence and electrical noise. Two-dimensional detector images were integrated around the position where the Bragg peak was observed. Each data point contains statistics from 300-600 shots. The 300 fs jitter of the free electron laser was corrected with a GaAs timing tool for delays $t < 7$ ps, yielding an effective time resolution of 150 fs at FWHM (37).

Figure 1B reports the magnetic Bragg peak ratio $I_{\text{on}}/I_{\text{off}}$, where $I_{\text{on}}$ denotes the intensity after laser excitation and $I_{\text{off}}$ when the laser was switched off. It is noted that even for strong fluxes some remnant magnetic signal is observed for $t > 0$, suggesting some experimental mismatch between pumped laser and probed x-ray volumes Further information on the time evolution of the magnetic order parameter is given in the SI Appendix.

Time-resolved resonant inelastic scattering (tr-RIXS). The ultrafast recovery of magnetic short-range correlations was studied at the X-ray Pump Probe (XPP) instrument of the Linac Coherent Light Source (LCLS) running at a repetition rate of 120 Hz (38). The data at $(\pi, \pi)$ and $(\pi, 0)$ correspond to measurements at (~3.5, 0.5, 2) and (~3.5, 0, 19.6) with $2\theta = 93.7$ and 91.8°. The x-rays were focused to a spot size of 50 μm FWHM. We used a grazing incidence geometry of 3.7 and 2.1° for the two reciprocal lattice positions, yielding an x-ray penetration depth of 378 and 214 nm, respectively (see Supplemental Information). The laser spot size equaled 845 μm FWHM at normal incidence and the fluence was fixed to 20 mJ/cm². The same RIXS spectrometer as in Ref. (2) was used, delivering an energy resolution of ~70 meV with an angular acceptance of 6° (0.28 Å⁻¹) on the analyser. The ePix100a area detector was read out every shot (39). The time-resolution of the experiment arising from a combined jitter and beam drift effect was limited to ~0.4 ps. It is noted that it proved impractical to use the timing tool, as it was insufficiently sensitive on the aggressively monochromatized incident beam. RIXS spectra were collected in a stationary mode of 144000 shots each. Two to six acquisitions were taken for each time delay. The data presented in Fig. 2A and B were normalized to an incident beam intensity monitor and calibrated to the energy of the orbital excitation at 680 meV (20–23). The center of the elastic line and its width were fixed in the analysis to 0 meV and the experimental resolution, respectively. Robust fits were obtained by further constraining the energy of the magnon continuum to 200 meV and its amplitude to the mean values of 2.02 and 1.44 for $(\pi, \pi)$ and $(\pi, 0)$, respectively. All other parameters were varied freely, and shown in the SI appendix, Fig. S2.

ACKNOWLEDGMENTS. We thank Chris Homes for discussions. The design, execution of the experiments, as well as the data analysis are supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Science and Engineering Division, under Contract No. DE-SC0012704(BNL) and DE-AC02-06CH11357(ANL). D.G.M. acknowledges funding from the Swiss National Science Foundation, Fellowship No. P2EZP2_175092. C.D.D. was supported by the Engineering and Physical Sciences Research Council (EPSRC) Centre for Doctoral Training in the Advanced Characterisation of Materials under Grant No. EP/L015277/1. X. L. and J.Q.L. were supported by the ShanghaiTech University startup fund, MOST of China under Grant No. 2016YFA0401000, NSFC under Grant No. 11304017 and the Chinese Academy of Sciences under Grant No. 112111KYSB20170050. Work at UCL was supported by the EPSRC under Grants No. EP/N027671/1 and No. EP/N034872/1. The work at ICFO received financial support from the Spanish Ministry of Economy and Competitiveness through the “Severo Ochoa” program for Centres of Excellence in R&D (SEV-2015-0522), from Fundació Privada Cellex, from Fundació Mir-Puig, and from Generalitat de Catalunya through the CERCA program and from the European Research Council (ERC) under the European Union’s Horizon 2020 research and innovation programme (Grant agreement No. 758461). J.L. acknowledges support from the National Science Foundation under Grant No. DMR-1848269. The magnetic Bragg peak measurements were performed at the BL3 of SACLA with the approval of the Japan Synchrotron Radiation Research Institute (JASRI) (Proposal No. 2017A8077, 2018A8032 and 2019A8087). This research made use of the Linac Coherent Light Source (LCLS), SLAC National Accelerator Laboratory, which is a DOE Office of Science User Facility, under Contract No. DE-AC02-07SF00515.


I. SI TIME-RESOLVED RESONANT ELASTIC X-RAY SCATTERING RESULTS

The response of magnetic long-range order on the laser pump was first investigated via reciprocal lattice scans along the (0, 0, L) and (H, 0, 0) directions around the (-3.5, 1.5, 18) magnetic Bragg peak [notation in reciprocal lattice units (r.l.u)]. Figure S1 displays datasets before and \(\sim 2.5\) ps after the arrival of a 32.2 mJ/cm\(^2\) strong laser pulse. The laser radiation induces a suppression, but no broadening, of the magnetic Bragg peak within our experimental \(Q\)-resolution. This provides evidence that all crucial information on transient magnetic long-range order is captured by the evolution of the magnetic Bragg peak intensity. It is noted that additional \(H\)- and \(L\)-scans were taken \(\sim 40\) ps after laser excitation confirming this conclusion.

Figure S2A shows the (-3.5, 1.5, 18) magnetic Bragg signal on the area detector before and around 1 ps after the arrival of the laser pulse. The time evolution of the depleted magnetic Bragg peak intensity is shown in the upper panel of Fig. S2B. Immediately after the optical pump \((t = 0)\) magnetism is reduced by 50-75% for laser fluences between 5.2 and 32.2 mJ/cm\(^2\). A significant fraction of the magnetic signal is recovered within the first picoseconds, but a slower process is superimposed preventing the full restoration of the original magnetic state. As a result, the reduced magnetic peak intensity stays roughly constant in the range between 5 and 500 ps without noticeable recovery. Furthermore, it is noted that a faint oscillatory behavior appears \(\sim 50\) ps after laser impact for fluences larger than 14.2 mJ/cm\(^2\). This feature is a typical signature of a strain wave that is propagating through the crystal and subsequently reflected at the sample boundary [1–3].

The time evolution of the transient magnetic long-range order is described by the minimal phenomenological model

\[
I(t) = \begin{cases} 
1 - Ae^{-t/\tau_{\text{mag}}} - C(1 - e^{-t/\tau_{\text{mag}}}) + De^{-t/\tau_{\text{wave}}} \sin(\omega t), & \text{for } t \geq 0 \\
1, & \text{for } t < 0
\end{cases},
\]

where \(A\) quantifies a prompt, step-like decay at \(t = 0\) (as it is much faster than the time resolution), which recovers on a timescale \(\tau_{\text{mag}}\), and \(C\) is the fraction of the order that has not been restored in 500 ps. The strain wave features an amplitude \(D\), decay time \(\tau_{\text{wave}}\) and oscillation frequency \(\omega\). The measured magnetic Bragg peak intensity is modeled by convolving \(I(t)\) with a Gaussian function in order to account for our finite time resolution of 0.15 ps (full-width at half-maximum). The initial decay of magnetic long-range order was found to be much faster than the time resolution, and was thus approximated as an instantaneous step function in Eq. 1.

Robust fits of experimental results were obtained by constraining the amplitude of the strain wave to \(D = 0.5\). The frequency of the strain wave at 23.3 mJ/cm\(^2\) was fixed to 0.03 rad/s (the mean value of all other conditions) to
FIG. S2. Time evolution of magnetic order in Sr$_3$Ir$_2$O$_7$. (A) Intensity of the (-3.5, 1.5, 18) magnetic Bragg peak [notation in reciprocal lattice units (r.l.u)] in equilibrium (top) and after laser radiation of ∼20 mJ/cm$^2$ (bottom). The detector pixels are 0.0086 r.l.u wide (B) Top panel: Relative magnetic Bragg peak intensity as a function of time delay. The error bars follow Poissonian statistics. Panel is split into short (6 ps) and long (100 ps) time ranges for clarity. Bottom panel: 14.2 mJ/cm$^2$ Sr$_3$Ir$_2$O$_7$ data normalized to the maximal depleted volume fraction and fit with the model shown in Eq. 1. The results are compared to data for Sr$_2$IrO$_4$ taken from Ref. [4] with a fluence chosen to match the fast recovery of the magnetic volume fraction. Heisenberg-like Sr$_2$IrO$_4$ reveals a slow recovery with onset around 100 ps, which is absent in gapped Sr$_3$Ir$_2$O$_7$. (C) Fluence dependence of the persistent transient fraction, $C$, and the fast recovery timescale $\tau_{\text{mag}}$. The gray area shows the standard deviation around the statistical mean value of the fast recovery timescale, i.e. $\tau_{\text{mag}} = 1.3(3)$ ps.

further stabilize the fit. All other parameters were varied freely and shown in Tab. 1. It is noted that even for strong fluences $A < 1$, suggesting some experimental mismatch between pumped laser and probed x-ray volumes (see also below). This emphasizes the need to also include contributions like the energy transfer out of the illuminated sample volume in theoretical approaches that model transient magnetic states.

The lower panel of Fig. S2B displays the best fit to the normalized intensity at 14.2 mJ/cm$^2$ (see also Materials and Methods section). The fluence dependence of $\tau_{\text{mag}}$ and $C$ is displayed in Fig. S2C. The fast timescale $\tau_{\text{mag}} = 1.3(3)$ ps is fluence independent and we find $C > 0$ for all fluences. This is different than in nearly gapless Sr$_2$IrO$_4$ - a related material where at fluences leading to a similar fast recovery of the magnetic volume fraction, the onset of a slow magnetic recovery is observed around 100 ps (see lower panel of Fig. S2B) [4]. The different behaviors are likely to arise from their distinct magnetic interactions and resulting magnon dispersions. A phonon-assisted energy transfer from excited (pseudo-)spins into the lattice degrees of freedom, for instance, is expected to be reduced in large gap antiferromagnets, yielding a much slower magnetic recovery when compared to nearly gapless materials.

<table>
<thead>
<tr>
<th>fluence (mJ/cm$^2$)</th>
<th>$A$</th>
<th>$C$</th>
<th>$\tau_{\text{mag}}$ (ps)</th>
<th>$\tau_{\text{wave}}$ (ps)</th>
<th>$\omega$ (rad/s)</th>
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<td>0.54(4)</td>
<td>0.023(5)</td>
<td>0.85(5)</td>
<td>NaN</td>
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<td>0.66(3)</td>
<td>0.154(4)</td>
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<td>12(2)</td>
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<td>14.2</td>
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<td>0.225(3)</td>
<td>1.58(9)</td>
<td>11(1)</td>
<td>0.035(4)</td>
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<td>18.7</td>
<td>0.72(4)</td>
<td>0.303(4)</td>
<td>1.26(9)</td>
<td>18(2)</td>
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<td>0.71(2)</td>
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<td>1.7(1)</td>
<td>19(1)</td>
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<tr>
<td>32.2</td>
<td>0.78(2)</td>
<td>0.429(4)</td>
<td>1.3(1)</td>
<td>13(1)</td>
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II. SI TIME-RESOLVED RESONANT INELASTIC X-RAY SCATTING RESULTS

The time-resolved resonant inelastic x-ray scattering (tr-RIXS) spectra in Fig. 2a and b of the main manuscript reveal an elastic line, a collective magnon at \(\sim 100\) and 150 meV for \((\pi, \pi)\) and \((\pi, 0)\), respectively, a magnon continuum at \(\sim 250\) meV and an orbital excitation at 680 meV. A sum of four Gaussian-shaped peaks was used to represent the energy-loss data. The center of the elastic line and its width were fixed in the analysis to 0 meV and the experimental resolution of 70 meV, respectively. Robust fits were obtained by further constraining the energy of the magnon continuum to 260 meV and its amplitude to the mean free value of 2.02 and 1.44 for \((\pi, \pi)\) and \((\pi, 0)\), respectively. All other parameters were varied freely. Figure S3 shows the relative time dependence of the integrated magnon, \(I_{\text{mag}}\), and orbital, \(I_{\text{dd}}\), intensity, the magnon and orbital position, \(E_{\text{mag}}\) and \(E_{\text{dd}}\), the amplitude of the elastic line, \(A_{\text{el}}\), and the full-width at half-maximum of the broad magnetic feature \(\Gamma_{\text{Bi-mag}}\) at higher energies.

III. SI ESTIMATION OF THE X-RAY AND LASER PENETRATION DEPTH

The optical penetration depth \(\Lambda\) of a beam incident at angle \(\alpha\) with respect to the sample surface is defined by [5]

\[
\Lambda = \frac{\lambda}{4\pi} \left[ \Im \left( \sqrt{N^2 - \cos^2(\alpha)} \right) \right]^{-1},
\]

where \(\Im\) signifies the imaginary part, \(\lambda\) is radiation wavelength, and \(\tilde{N} = n + ik\) is the complex index of refraction. We define the penetration depth as the distance over which the beam intensity drops by \(1/e\), and we remind the reader that \(\tilde{N} = \sqrt{\varepsilon}\) where \(\varepsilon\) is the relative permittivity. For the x-ray calculation, the dielectric constants below and above the Ir \(L_3\)-edge were calculated based on the crystal structure of \(\text{Sr}_3\text{Ir}_2\text{O}_7\) using the xrayutilities Python package [6]. The imaginary part of the index of refraction at the resonance was corrected considering the x-ray absorption white-line intensity [7]. At the Ir \(L_3\)-edge we find \(\Lambda_{\text{x-ray}} = 162, 214\) and 378 nm for \(\alpha = 1.6, 2.1\) and 3.7°, respectively. Using the reported optical conductivity results of Ref. [8], we find \(\Lambda_{\text{laser}} \approx 82\) nm for \(\alpha \approx 20°\).
IV. SI SPIN-WAVE DISPERSION AND ESTIMATION OF NÉEL TEMPERATURE

The superexchange interactions in octahedrally coordinated 5d$^5$ iridates are strongly dependent on bond angles. In general, isotropic Heisenberg, anisotropic Heisenberg, Dzyaloshinskii-Moriya and Kitaev exchange couplings can be present. For the simplified case of exactly straight bonds, symmetric octahedra coordination and assuming that Coulomb repulsion, $U$, is much larger than Hund’s exchange, $J_H$, pseudo-dipolar and Kitaev interactions vanish by symmetry and the magnetic exchange becomes purely isotropic Heisenberg-like [9]. Since Sr$_2$IrO$_4$ and Sr$_3$Ir$_2$O$_7$ possess iodium-oxygen-iridium bond angles that are close to 180° [10–12], the dominant exchange term is Heisenberg-like, and the subdominant terms are anisotropic Heisenberg, Dzyaloshinskii-Moriya and Kitaev exchanges with the latter term generally considered negligible. This has been predicted in several theoretical works [13–15] and verified by RIXS and Raman scattering [16–18]. Thus, magnetism in Sr$_2$IrO$_4$ can be described by a Heisenberg Hamiltonian with in-plane isotropic coupling $J$, anisotropic coupling $\Gamma$, Dzyaloshinskii-Moriya interaction $D$ and interlayer coupling $J_c$ [13]

$$H_{214} = \sum_{\langle \vec{n}, \vec{m} \rangle} \left[ J_{n+} S_{\vec{n}, \vec{m}}^z + \Gamma S_{\vec{n}, \vec{m}}^z S_{\vec{m}, \vec{m}}^z + D(-1)^{n_x+n_y}(S_{\vec{n}, \vec{m}}^x S_{\vec{m}, \vec{m}}^y - S_{\vec{n}, \vec{m}}^y S_{\vec{m}, \vec{m}}^x) \right] + \sum_{\langle \langle \langle \vec{n}, \vec{m} \rangle \rangle \rangle} J_3 S_{\vec{n}, \vec{m}}^z S_{\vec{m}, \vec{m}}^z + \sum_{\vec{n}, \vec{m}} J_{\vec{n}, \vec{m}} S_{\vec{n}, \vec{m}}^z S_{\vec{m}, \vec{m}}^+ S_{\vec{m}, \vec{m}}^-. $$

(3)

Here, $\vec{n} = (n_x, n_y)$ are vectors to sites within the IrO$_2$ layers and $l$ is the layer index, defining $S_{\vec{n}, l}$ as the spin operator at site $\vec{n}$ of layer $l$. $\langle \langle \langle \vec{n}, \vec{m} \rangle \rangle \rangle$ and $\langle \langle \langle \vec{n}, \vec{m} \rangle \rangle \rangle$ denote first, second and third nearest neighbors in the tetragonal plane and $J$, $J_2$ and $J_3$ are the corresponding isotropic interaction constants. We mention that some reported models also include an anisotropic, symmetric exchange term in $H_{214}$ [14, 15]. This term is, however, much smaller than the interlayer coupling and can thus, be neglected for our purposes.

In cases where the spins are aligned in the tetragonal plane, the nearest-neighbor interactions in Eq. 3 can be mapped onto an effective isotropic exchange Hamiltonian with interlayer coupling as [13]:

$$H_{iso} = \sum_{\langle \vec{n}, \vec{m} \rangle} J \vec{S}_{\vec{n}, l} \vec{S}_{\vec{m}, l} + \sum_{\vec{n}, \vec{m}} J_c \vec{S}_{\vec{n}, l} \vec{S}_{\vec{m}, l+1}. $$

(4)

The transformation involves a rotation of the two sublattices in the tetragonal plane, where $\vec{S}_{\vec{n}, l}$ is the rotated spin operator and $\vec{J} = \sqrt{J^2 + D^2}$. Since the transition temperature in the case of $J \gg J_c$ is dominated by the spin-wave velocity, we have neglected the second and third nearest neighbor couplings here. $T_N$ is defined as the temperature for which the expectation value $\langle \vec{S}_{\vec{n}}^z \rangle$ (calculated with linear spin-wave theory) vanishes. For $T_N \ll J/2$ the Néel temperature is given by [19]

$$T_N = \frac{\pi \vec{J}}{\log(T_\text{iso}/J_c)}. $$

(5)

So if the interlayer coupling in Sr$_2$IrO$_4$ could be switched off, it would be predicted not to display magnetic order at any finite temperature. Using $\vec{J} = 50$ meV to match a spin-wave energy of 200 meV at $(\pi, 0)$, we can reproduce the experimentally observed $T_N = 285$ K by assuming an interlayer coupling of 1.1 $\mu$eV. $T_N = 384.2$ K is obtained when using the previously reported value $J_c = 15.9$ $\mu$eV [13].

As c-axis coupling gain particular importance in Sr$_3$Ir$_2$O$_7$ the Hamiltonian of Eq. 3 needs to be extended to

$$H_{327} = \sum_{\langle \vec{n}, \vec{m} \rangle, l} \left[ J \vec{S}_{\vec{n}, l} \vec{S}_{\vec{m}, l} + \Gamma S_{\vec{n}, l} S_{\vec{m}, l}^z \right] + \sum_{\langle \langle \langle \vec{n}, \vec{m} \rangle \rangle \rangle} D(-1)^{n_x+n_y+l}(S_{\vec{n}, l}^x S_{\vec{m}, l}^y - S_{\vec{n}, l}^y S_{\vec{m}, l}^x)$$

$$\sum_{\vec{n}} \left[ J_c \vec{S}_{\vec{n}, 1} \vec{S}_{\vec{n}, 2} + \Gamma c S_{\vec{n}, 1} S_{\vec{n}, 2}^z \right] + \sum_{\vec{n}} D_c(-1)^{n_x+n_y}(S_{\vec{n}, 1}^x S_{\vec{n}, 2}^y - S_{\vec{n}, 1}^y S_{\vec{n}, 2}^x) $$

(6)

with the additional terms $\Gamma_c$ and $D_c$, which couple the planes within a bilayer. This model neglects coupling between the bilayers. The Néel temperature of a collinear antiferromagnetic spin alignment is found from the numerical solution of $\langle \vec{S}_{\vec{n}, l}^z \rangle = 0$ with

$$\langle \vec{S}_{\vec{n}, l}^z \rangle = 1 - \frac{1}{16\pi^2} \int \left[ \frac{B_+}{E_+(\vec{q})} \coth(\beta E_+(\vec{q})) + \frac{B_-}{E_-(\vec{q})} \coth(\beta E_-(\vec{q})) \right] d\vec{q} d\vec{q}. $$

(7)
\[ E_{\pm}(\vec{q}) = \sqrt{B_{\vec{q},\pm}^2 - \left| C_{\vec{q},\pm} \right|^2} \] define the two magnon branches with

\[ \beta = 1/(k_B T_N) \]
\[ B_{\vec{q},\pm} = \frac{1}{2}(8J + 8\Gamma + J_c + \Gamma_c) - 4J_2(1 - \cos(q_x) \cos(q_y)) - 4J_3(1 - \gamma_2q) - 2J_2c(1 \mp \gamma q) \]
\[ C_{\vec{q},\pm} = \frac{1}{2}(8J_\mp \mp q \pm J_c) - \frac{i}{2}(8D_{\vec{q}} \mp D_c) \]
\[ \gamma_q = \frac{1}{2}(\cos(q_x) + \cos(q_y)) \]

and \( k_B = 8.617 \times 10^{-2} \text{ meV/K} \) the Boltzmann constant. A reasonable Néel temperature \( T_N = 118 \text{ K} \), which is within a factor of three of the experimental value, is obtained for \( J = 46.6 \text{ meV}, J_c = 25.2 \text{ meV}, J_2 = 5.95 \text{ meV}, J_3 = 7.3 \text{ meV}, J_2c = 6.2 \text{ meV}, \Gamma = 2.2 \text{ meV}, \Gamma_c = 34.3 \text{ meV}, D = 12.25 \text{ meV} \) and \( D_c = 28.1 \text{ meV} \). The analysis shows that magnetic order perpendicular to the tetragonal plane can be stabilized even in the absence of long-range c-axis coupling. Furthermore, in \( \text{Sr}_3\text{Ir}_2\text{O}_7 \) we do not expect the same extreme anisotropy between the ab-plane and c-axis correlations as in \( \text{Sr}_3\text{Ir}_2\text{O}_4 \).

8. G. Ahn, S. J. Song, H. S. Wilson, and S. J. Moon, Infrared spectroscopic evidences of strong electronic correlations in \( \text{Sr}_{1-x}\text{La}_x\text{Ir}_2\text{O}_7 \), Scientific Reports 6, 32632 (2016).
13. T. Takayama, A. Matsumoto, G. Jackeli, and H. Takagi, Model analysis of magnetic susceptibility of \( \text{Sr}_3\text{Ir}_4\text{O}_7 \): A two-dimensional \( J_{\text{eff}} = \frac{1}{2} \) Heisenberg system with competing interlayer couplings, Phys. Rev. B 94, 224420 (2016).