**Supplementary Information for**

**Giant stress response of terahertz magnons in a spin-orbit Mott insulator**

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Supplementary Note 1: Strain calibration using x-ray diffraction

Since experimental constraints imposed by the specific Raman setup used prevented us from x-ray diffraction measurements in-situ, the strain level was estimated during the Raman measurements using a built-in displacement sensor based on a parallel-plate capacitor, which has been proven to yield reasonable estimates in previous studies [1–6]. Nevertheless, subtle details such as the shape of the sample, the amount of epoxy used, and the epoxy thickness, can affect the accuracy of our strain estimation, thus making it difficult to precisely extract the parameters in our theory.

To obtain the best estimate of the strain present in the Raman experiments, we therefore performed separate x-ray diffraction measurements on the same sample mounted on the strain device to collect the Raman data. We controlled the sample strain by applying different voltages to the piezoelectric stacks of the strain device. This results in capacitance changes of the built-in displacement sensor, which is based on a parallel-plate capacitor. Using the inverse relation between the capacitance value and the displacement of a parallel-plate capacitor (\(C = \varepsilon_0 A/d\), where \(C\) is the capacitance, \(\varepsilon_0\) is the vacuum permittivity, \(A\) is the area of the parallel plates, and \(d\) is the distance between the plates), one obtains displacement changes for each strain step.

Figures S1(a),(c) show diffraction scans and Raman spectra as a function of displacement, respectively. From the x-ray measurements, we obtain a linear relation between the displacement and the lattice constant change, as shown in Figure S1(b). From the Raman scattering experiments, we extract a linear relation between the displacement and the energy of the phonon at 394 cm\(^{-1}\), as displayed in Figure S1(d). Combining these two results, we obtain a phonon energy change against uniaxial strain of \(-0.188/0.0444 = -4.23\) (cm\(^{-1}\)/%). This value, along with zero-strain references obtained from broken, fully relaxed samples after each strain experiment, was used to calibrate the strain scale of separate Raman experiments. The strain scale also includes the effect of the in-plane Poisson ratio of \(\nu = 0.3\), which is the typical value for transition metal oxides with perovskite structure.

Notice that the first application of a small external stress applied along the \([1 0 0]\) axis is only shifting twin domain boundaries without altering the intrinsic strain value \(\varepsilon_0\), until a single domain type out of the two degenerate ones is selected and occupies the entire sample volume. Only after that, the local orthorhombic distortion starts to increase as \(\varepsilon_0 + \varepsilon\), and one observes the magnon energy enhancement, as shown in Fig. 3(a) of the main text.

Supplementary Note 2: Interplay between spin-lattice coupling and spin-exchange anisotropy

In addition to the spin-lattice coupling mechanism, the in-plane magnon gap can also have contributions from the anisotropic spin exchange interactions. To describe this contribution on a phenomenological level, we introduce the following magnetic anisotropy energy \(E_{ex}\) of four-fold tetragonal symmetry:

\[
E_{ex} = -\frac{1}{2} \kappa S^2 \cos 4\alpha.
\]

A positive \(\kappa\) favors the easy axis along the Ir-Ir bond direction [110] (i.e. \(\alpha = 0\)), while \(\kappa < 0\) favors spin direction along the [100] diagonal (\(\alpha = \pi/4\)). Microscopically, the bond-directional pseudodipolar exchange interaction in Sr\(_2\)IrO\(_4\) generates exactly this type of anisotropy, with \(\kappa > 0\), via a quantum order-by-disorder mechanism. Having a quantum nature, this term should be weak compared to the spin-lattice mechanism that operates on a classical level, via a lattice symmetry-breaking phase transition. For generality, we consider both signs of \(\kappa\).
FIG. S1. Comparison between x-ray and Raman data under uniaxial strain. a, 2θ-scan of the (2 2 0) Bragg reflection measured at different displacement values. The solid lines are guides-to-the-eye. b, Lattice constant changes based on the shift of the (2 2 0) Bragg reflection as a function of displacement. The solid line is the result of a linear fit to the data. c, Raman spectra of the phonon at 394 cm$^{-1}$ measured at different displacement values. The solid lines are the results of Lorentzian fits to the data. d, Energy of the phonon at 394 cm$^{-1}$ as a function of displacement. The solid line is the result of a linear fit to the data.

The total anisotropy energy $E(\alpha) = E_{s-t} + E_{ex}$, including spin-lattice as well spin-exchange anisotropy, reads as:

$$E(\alpha) = \text{const} + \frac{1}{2} (\Gamma_{1} - \Gamma_{2} - \kappa) S^{2} \cos 4\alpha.$$  

In order to have the moments along the diagonal direction ($\alpha = \pi/4$) as in Sr$_{2}$IrO$_{4}$, the parameter $\kappa$ must obey the constraint $\kappa < \Gamma_{1} - \Gamma_{2}$.

The magnon gap at zero strain $\varepsilon = 0$ is thus determined by the combined effect of the spin-lattice and spin-exchange anisotropies:

$$\Delta(\varepsilon = 0) = 8S \sqrt{J(\Gamma_{1} - \kappa)} = \Delta_{0} \sqrt{1 - \frac{\kappa}{\Gamma_{1}}}.$$  

Here, $\Delta_{0} = 8S \sqrt{J\Gamma_{1}}$ is a magnon gap induced by spin-lattice contribution alone. Note that the pseudodipolar exchange anisotropy with $\kappa > 0$ reduces the gap value, as it favors spins along the Ir-Ir bond direction, thereby competing with the spin-lattice coupling (which dictates easy axis along [100]).
When the strain is applied along [1 0 0] direction, the $\kappa$ term does not affect a functional form of the gap, i.e. the magnon gap monotonically increases as a function of the external strain $\varepsilon$:

$$\Delta(\varepsilon) = \Delta_0 \sqrt{1 - \frac{\kappa}{\Gamma_1}} + \frac{|\varepsilon|}{\varepsilon_0} = \Delta(\varepsilon = 0) \sqrt{1 + \frac{|\varepsilon|}{\varepsilon_0}}.$$ \hspace{1cm} (4)

Here $\varepsilon_0 = \varepsilon_0(1 - \frac{\kappa}{\Gamma_1})$, and $\varepsilon_0 = 2S^2g_1/K_1$ is the spin-lattice induced spontaneous orthorhombic distortion. The external strain [100] does not change the easy axis direction but monotonically increases the orthorhombicity and the size of the magnon gap.

External strain applied along [1 1 0] direction rotates the orthorhombicity axis and spin direction towards the Ir-Ir bond direction $\alpha = 0$, which is reached at some value of $\varepsilon = \varepsilon_c$ (see Fig. 4 of the main text). In the spin-lattice coupling theory, this rotation does not affect the magnon gap, as it is only sensitive to the amplitude of the distortion but not to its direction. Above $\varepsilon_c$, the easy axis remains along the [110] direction, but the orthorhombic distortion and hence the magnon gap start to increase. The $\kappa$ term originating from spin-exchange anisotropy results in a new feature. Namely, the magnon gap becomes a function of strain even below $\varepsilon_c$:

$$\Delta(\varepsilon \leq \varepsilon_c) = \Delta_0 \sqrt{1 - \frac{\kappa}{\Gamma_1}} \left(1 - \varepsilon_c^2/\varepsilon_0^2\right). \hspace{1cm} (5)$$

Here, a critical strain value $\varepsilon_c = \varepsilon_0 \frac{g_1}{g_2} \left(1 - \frac{\Gamma_2}{\Gamma_1} - \frac{\kappa}{\Gamma_1}\right)$ is renormalized by $\kappa$. We note that at $\varepsilon = \varepsilon_c$ the magnon gap equals the spin-lattice contribution $\Delta_0$.

For the strain values $\varepsilon > \varepsilon_c$, we obtain a monotonical increase of the gap as in a pure spin-lattice coupling theory (cf. Eq. 8 of the main text):

$$\Delta(\varepsilon > \varepsilon_c) = \Delta_0 \sqrt{1 + \frac{g_2}{g_1} \frac{|\varepsilon| - \varepsilon_c}{\varepsilon_0}}.$$ \hspace{1cm} (6)

The above equations determine the magnon gaps as a function of the external strain $\varepsilon$; the results for different $\kappa$ values are shown in Fig. S2. The regime change in the magnon gap behavior at $\varepsilon_c$ is preserved, but the kink at this point varies with $\kappa$. We see that the negative $\kappa$ dependences are qualitatively different from the observed behavior and thus can be excluded. This is expected as the pseudodipolar exchange interaction results in $\kappa > 0$.

To quantify the value of $\kappa$ in Sr$_2$IrO$_4$, we use the above equations to fit the experimental data. The results for $\kappa/\Gamma_1 = 0.03$, shown in Fig. S3, well reproduce the data, including a variation of the gap at small strain values $\varepsilon_{110} < \varepsilon_c$. We see that while the spin-exchange anisotropy term $\kappa$ may improve a detailed comparison with experiment,
FIG. S3. Fitting the experimental data including the spin-exchange anisotropy $\kappa$ term. The parameters used are $\kappa/\Gamma_1 = 0.03$, $\varepsilon_0 = 1.1 \times 10^{-3}$, $\varepsilon_c = 3 \times 10^{-4}$, and $g_2/g_1 = 1.1$.

it is actually very weak in Sr$_2$IrO$_4$ (just about a 0.03 fraction of the spin-lattice coupling energy $\Gamma_1$). This is not surprising because the exchange Hamiltonian in Sr$_2$IrO$_4$ is dominated by the isotropic Heisenberg coupling. Moreover, the anisotropic exchange terms like pseudodipolar coupling contribute to the in-plane magnetic anisotropy only via quantum (order-by-disorder) effects, further reducing their impact on the magnon gaps. We thus conclude that the spin-lattice coupling is the major source of the magnetic anisotropy in Sr$_2$IrO$_4$, as we considered in the main text.

[1] Hicks, C. W., Barber, M. E., Edkins, S. D., Brodsky, D. O. & Mackenzie, A. P. Piezoelectric-based apparatus for strain tuning. Rev. Sci. Instrum. 85, 065003 (2014).
[2] Hicks, C. W. et al. Strong Increase of $T_c$ of Sr$_2$RuO$_4$ Under Both Tensile and Compressive Strain. Science 344, 283 (2014).
[3] Steppke, A. et al. Strong peak in $T_c$ of Sr$_2$RuO$_4$ under uniaxial pressure. Science 355, 9398 (2017).
[4] Barber, M. E., Gibbs, A. S., Maeno, Y., Mackenzie, A. P. & Hicks, C. W. Resistivity in the Vicinity of a van Hove Singularity: Sr$_2$RuO$_4$ under Uniaxial Pressure. Phys. Rev. Lett. 120, 076602 (2018).
[5] Kim, H.-H. et al. Uniaxial pressure control of competing orders in a high-temperature superconductor. Science 362, 1040 (2018).
[6] Kim, H.-H. et al. Charge Density Waves in YBa$_2$Cu$_3$O$_{6.67}$ Probed by Resonant X-Ray Scattering under Uniaxial Compression. Phys. Rev. Lett. 126, 037002 (2021).