Raman spectroscopic signature of fractionalized excitations in the harmonic-honeycomb iridates $\beta$- and $\gamma$-Li$_2$IrO$_3$

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The fractionalization of elementary excitations in quantum spin systems is a central theme in current condensed matter physics. The Kitaev honeycomb spin model provides a prominent example of exotic fractionalized quasiparticles, composed of itinerant Majorana fermions and gapped gauge fluxes. However, identification of the Majorana fermions in a three-dimensional honeycomb lattice remains elusive. Here we report spectroscopic signatures of fractional excitations in the harmonic-honeycomb iridates $\beta$- and $\gamma$-Li$_2$IrO$_3$. Using polarization-resolved Raman spectroscopy, we find that the dynamical Raman response of $\beta$- and $\gamma$-Li$_2$IrO$_3$ features a broad scattering continuum with distinct polarization and composition dependence. The temperature dependence of the Raman spectral weight is dominated by the thermal damping of fermionic excitations. These results suggest the emergence of Majorana fermions from spin fractionalization in a three-dimensional Kitaev–Heisenberg system.
The fractionalization of elementary excitations is a characteristic feature of quantum spin liquids. Such a liquid evades conventional magnetic order even at $T = 0$ K and thereby preserves all symmetries of the underlying spin Hamiltonian. In the last decade, there has been significant progress in the experimental identification of quantum spin liquids in a class of geometrically frustrated Heisenberg magnets with elementary excitations that are given by chargeless spinons carrying spin $s = 1/2$. For two-dimensional (2D) triangular and Kagome lattices, however, a quantitative understanding of spinons remains unsatisfactory due to a lack of reliable theoretical methods of handling macroscopic degenerate ground states. In contrast, only a subtle Raman response and a high-energy Majorana excitation in fractionalization through a continuum-like excitation in the spin fractionalization in a 3D honeycomb lattice.

Results

Polarization dependence of Raman spectra. Figure 1a shows the crystal structures of $\beta$- and $\gamma$-Li$_2$IrO$_3$. $\beta$-Li$_2$IrO$_3$ consists of the zigzag chains (blue and orange sticks), which alternate in orientation between the two basal plane diagonals and are connected via the bridging bonds (green stick) along the $c$ axis. In $\gamma$-Li$_2$IrO$_3$, two interlaced honeycomb layers alternate along the $c$ axis. Figure 1b,c presents the polarization-dependent Raman responses $\chi'(\omega)$ of $\beta$- and $\gamma$-Li$_2$IrO$_3$ measured at $T = 6$ K in two different scattering geometries. Here the notation $(xy)$ with $x = a$ and $y = b$, $c$ refers to the incident and scattered light polarizations, which are parallel to the crystalline $x$ and $y$ axis, respectively. $\chi'(\omega)$ presents the dynamical properties of collective excitations and is obtained from the raw Raman spectra $I(\omega)$ using the relation $I(\omega) \propto [1 + n(\omega)]\chi'(\omega)$ where $1 + n(\omega) = 1/(1 - e^{-\hbar\omega/k_B T})$ is the Bose thermal factor.

Within the Fleury–Louden–Elliott theory, the magnetic scattering intensity of a 3D Kitaev system is given by the density of states of a weighted two-Majorana spinon, $I(\omega) = \pi \sum_{m,n,k} \delta(\omega - \epsilon_{m,k} - \epsilon_{n,k}) |B_{m,n,k}|^2$, where $\epsilon_{m,k}$ is a Majorana spinon dispersion with the band indices $m$, $n = 1,2,3$ for $\beta(\gamma)$-Li$_2$IrO$_3$ and $B_{m,n,k}$ is the matrix element creating two Majorana excitations. The observed $\chi'(\omega)$ is composed of sharp phonon excitations superimposed on a broad, featureless continuum extending up to $200$ meV. The Raman-active phonon modes are presented in Supplementary Figure 1 and Supplementary Tables 1 and 2 (see also Supplementary Note 1 for details). The magnetic continuum arises mainly from two-Majorana spinon excitations. This assignment is analogue to observations in the 2D honeycomb lattice $\alpha$-RuCl$_3$, in which a broad continuum is taken as evidence of fractionalized excitations. The striking similarity of the magnetic response between $\alpha$-RuCl$_3$ and $\beta$- and $\gamma$-Li$_2$IrO$_3$ suggests that the 3D honeycomb iridates and the 2D honeycomb ruthenate realize Kitaev magnetism to a similar extent.

To the multiple spinon bands in the 3D harmonic honeycomb system, the Raman response of $\beta$- and $\gamma$-Li$_2$IrO$_3$ will be polarization- and composition-dependent, emulating a number of band edges and van Hove singularities. As seen in Fig. 1b,c, the iridate compounds show commonly an asymmetric magnetic response towards lower energy. The polarization dependence is mostly evident in the $\omega$-dependence of $\chi'(\omega)$. Compared with $\chi'(ac)$, $\chi'(ab)$ with green shading becomes systematically suppressed as $\omega \rightarrow 0$. Examining its composition dependence, $\chi'(ac)$ of $\beta$- and $\gamma$-Li$_2$IrO$_3$ is plotted together in Fig. 1d after subtracting phonon modes. $\chi'(ac)$ of $\beta$-Li$_2$IrO$_3$ shows a round maximum at $\sim 33$ meV, whereas its spectral weight is depressed to zero as $\omega \rightarrow 0$. In contrast, $\chi'(ac)$ of $\gamma$-Li$_2$IrO$_3$ has two maxima at 26 and 102 meV along with a finite excitation gap of $\Delta = 5 - 6$ meV marked by the arrows in Fig. 1c,d. Here, the extracted gap is estimated by a linear extrapolation of $\chi'(\omega)$. The slightly richer spectrum of $\gamma$-Li$_2$IrO$_3$ than $\beta$-Li$_2$IrO$_3$ is linked to the increasing number of Majorana spinon bands. Thus, these results establish a subtle yet discernible polarization and composition dependence of $\chi'(\omega)$ in the 3D hyperhoneycomb compounds.

A related question is to what extent the hyperhoneycomb iridate materials retain the characteristic of Majorana fermions inherent to the 3D Kitaev model. For this purpose, we first compare the experimental and theoretical Raman response of $\beta$-Li$_2$IrO$_3$, which lies at the near-isotropic point with $J^3 = \mathcal{F} \approx \mathcal{F}^3$.
incarnadine shading. A magnetic continuum in (along the b axis. 

Similar trends are observed in the polarization dependence of the scattering intensity; the (ab) polarization spectrum has a much stronger intensity than the (ac) polarization spectrum, being in line with the theoretical calculations\(^8\). However, the low-energy spectrum does not open an excitation gap in the (ab) scattering channel and the fine spectral features anticipated in the bare two-Majorana spinon density of states do not show up. There is not much difference in the polarization dependence for the case of \(J_{z}\) - Li\(_2\)IrO\(_3\), which possesses three Majorana spinon bands and is at the anisotropic point with \(J_a \neq J_c \neq J_b\) (see Supplementary Fig. 2 and Supplementary Note 2 for the local bond geometry). The absence of the sharp spectral features and polarization-dependent spectral weights is ascribed to the unwanted spin-exchange terms including Heisenberg, off-diagonal and longer-range interactions. These subdominant terms on the one hand lead to a weak confinement of Majorana spinons, rendering the smearing out of the van-Hove singularities and the softening of spectral weight. On the other hand, they give rise to a bosonic (magnon) contribution to the magnetic continuum at low energies. In this regard, the excitation gap in \(J_{z}\) - Li\(_2\)IrO\(_3\) corresponds to an energy gap in the low-energy spin waves. As the pseudospin \(s = 1/2\) has a negligible single ion anisotropy, the anisotropic Kitaev interactions of \(J_{z}\)-IrO\(_3\) are responsible for opening the large gap. Notably, no obvious energy gap is present in the low-energy excitations of \(J_{z}\)-IrO\(_3\) with nearly isotropic Kitaev interactions.

Before proceeding, we estimate the Kitaev exchange interaction \(J_{z} = 17\) meV from the upper cutoff energy of the magnetic continuum. The extracted value is almost two times bigger than \(J_z = 8\) meV of \(\chi\)-RuCl\(_3\) (ref. 15), being consistent with larger spatial extent of Ir orbitals.

**Evolution of fermionic excitations.** The temperature dependence of the Raman spectra was measured for both \(J_{z}\) and \(\gamma\)-Li\(_2\)IrO\(_3\) in the (ac) and (cc) scattering symmetries, respectively. The representative spectra are shown in Fig. 2a,b. The broad magnetic continuum marked with pink shading develops progressively into a quasi-elastic response at low energies on heating through \(T_N\). The low-energy magnetic scattering grows more rapidly in \(J_{z}\) than \(\gamma\)-Li\(_2\)IrO\(_3\), because the latter has the large excitation gap. The magnetic Raman scattering at finite temperatures arises from dynamical spin fluctuations in a quantum paramagnetic state and can provide a good measure of the thermal fractionalization of quantum spins. The integrated Raman intensity in the energy range of \(1.5 J_z < h\omega < 3 J_z\) is plotted as a function of temperature in Fig. 2c,d. The temperature dependence of the integrated \(I(\omega)\) is well fitted by a sum of the Bose and the two-fermion scattering contribution \((1 - f(\omega))^2\) with the Fermi distribution function \(f(\omega) = 1/(1 + e^{\hbar\omega/k_B T})\) (ref. 35). The Bose contribution describes bosonic excitations such as magnons, whereas the two-fermion contribution is related to the creation or annihilation of pairs of fermions. The deduced energy \(h\omega = 0.76 - 0.79 J_z\) of fermions for \(J_{z}\) and \(\gamma\)-Li\(_2\)IrO\(_3\) validates the fitting procedure adopting a Fermi distribution function. Here we stress that the thermal fluctuations of fractionalized fermionic excitations are a Raman spectroscopic evidence of proximity to a Kitaev spin liquid. Essentially the same fermionic excitations were inferred from the \(T\)-dependence of the integrated spectral weight in \(\chi\)-RuCl\(_3\) (ref. 35).

Figure 2e,f shows the Raman conductivity \(\chi''(\omega)/\omega\) versus temperature. The Raman conductivity features a pronounced peak centred at \(\omega = 0\). The low-energy Raman response exhibits a strong enhancement with increasing temperature. The intermediate-to-high energy \(\chi''(\omega)/\omega\) above 30 meV dampens hardly with temperature. From the Raman conductivity we can
define a dynamic Raman susceptibility using Kramers–Kronig relation \( \chi^{\text{dyn}}(\omega) = \lim_{\omega \rightarrow 0} \alpha(k = 0, \omega) \equiv \frac{1}{\pi} \int_{0}^{\infty} \chi^{\text{static}}(\omega) \frac{d\omega}{\omega} \), that is, by first extrapolating the data from the lowest energy measured down to 0 meV and then integrating up to 200 meV. It is noteworthy to mention that \( \chi^{\text{dyn}} \) is in the dynamic limit of \( \chi^{\text{static}} \). Figure 2g\,h plots the temperature dependence of \( \chi^{\text{dyn}}(T) \) for \( \beta- \) and \( \gamma-\text{Li}_{2}\text{IrO}_{3} \). Irrespective of polarization and composition, \( \chi^{\text{dyn}}(T) \) shows a similar variation with temperature. On heating above \( T_{N} \), \( \chi^{\text{dyn}}(T) \) increases rapidly and then saturates for temperatures above \( T = 220 \text{–} 260 \text{ K} \). Remarkably, the energy corresponding to \( T^{*} \) is comparable to the Kitaev exchange interaction of \( J_{K} = 17 \text{ meV} \). We further note that the 2D Heisenberg–Kitaev material \( x\text{-RuCl}_{3} \) exhibits also a drastic change of magnetic dynamics through \( T \sim T_{K} = 100 \text{–} 140 \text{ K} \) (ref. 15). For temperatures below \( T^{*} \), the power law gives a reasonable description of \( \chi^{\text{dyn}}(T) \sim T^{\alpha} \), with \( \alpha = 1.58 \pm 0.05 \) and \( 2.64 \pm 0.09 \) in the respective (cc) and (ab) polarization for \( \beta-\text{Li}_{2}\text{IrO}_{3} \), and \( \alpha = 1.77 \pm 0.06 \) for \( \gamma-\text{Li}_{2}\text{IrO}_{3} \). As discussed in Supplementary Fig. 3 and Supplementary Note 3, \( \chi^{\text{dyn}}(T) \) is temperature independent in the paramagnetic phase as paramagnetic spins are uncorrelated. This is contrasted to the power-law dependence of \( \chi^{\text{dyn}}(T) \) in a spin liquid. This power-law is associated with slowly decaying correlations inherent to a spin liquid and the onset temperature \( T^{*} \) heralds a thermal fractionalization of Kitaev spins.

We now compare the dynamic Raman susceptibility with the static spin susceptibility given by SQUID magnetometry. As evident from Fig. 2g\,h, they behave in an opposite way. This discrepancy indicates that a large number of correlated spins are present in the limit \( \omega \rightarrow 0 \).

Fano resonance of optical phonon and magnetic specific heat. The phonon Raman spectra unveil a strongly asymmetric lineshape at 24 meV in \( \beta-\text{Li}_{2}\text{IrO}_{3} \) (see Fig. 3a) that is well fitted by a Fano profile \( I(\omega) = I_{0}(\omega + c)^{2} / (1 + c^{2}) \) (ref. 38). The reduced energy is defined by \( \epsilon = (\omega - \omega_{0}) / \Gamma \) where \( \omega_{0} \) is the bare phonon frequency, \( \Gamma \) the linewidth and \( q \) the asymmetry parameter. In Fig. 3b\,c, we plot the resulting frequency shift, the linewidth and the Fano asymmetry as a function of temperature. The errors are within a symbol size. Based on lattice dynamical calculations (see Supplementary Note 1), this phonon is assigned to an \( A_{g} \) symmetry mode, which involves contracting vibrations of Ir atoms along the \( c \) axis (see the sketch in the inset of Fig. 3a). Therefore, the observed anomalies could shed some light on the thermal evolution of Kitaev physics, because a Fano resonance has its root in strong coupling of phonons to a continuum of excitations.

With decreasing temperature, the Fano asymmetry, \( 1/|q| \), increases continuously and then becomes constant below the magnetic ordering temperature. As clearly seen from Fig. 3b, the temperature dependence of \( 1/|q| \) follows the two-fermion scattering form \( (1 - f(\omega))^{2} \), which gives a nice description of the temperature dependence of the integrated \( I(\omega) \) (see Fig. 2c,d). It is striking that the magnitude of the Fano asymmetry parallels a thermal damping of the fermionic excitations. In a Kitaev honeycomb system, spins are thermally fractionalized into the itinerant Majorana spinons. As a result, the continuum stemming from the spin fractionalization strongly couples to lattice vibrations that mediate the Kitaev interaction. It is noteworthy that the 24 meV mode involves the contracting motion of the bridging bonds between consecutive zigzag chains along the \( c \) axis. In addition, \( x\text{-RuCl}_{3} \) shows a Fano resonance of a phonon, which reinforces our assertion that the Fano asymmetry is an indicator of the thermal fractionalization of spins into the Majorana fermions.

As the temperature is lowered, phonon modes usually increase in energy and narrow in linewidth due to a suppression of anharmonic phonon–phonon interactions. Indeed, as shown in
In addition, the predicted topological transition at is somewhat higher than that of the theoretical value of 0.6 \w Fano asymmetry |\lambda| \times \text{ lattice temperature of the magnetic ordering. Unlike the 2D honeycomb Supplementary Note 4). A small kink in described by conventional anharmonic decay processes (see also profile. The inset depicts a schematic representation of eigenvector of the 24 meV temperature-dependent magnetic background. The 21 meV phonon on a low-energy side of the Fano resonance is fitted together with a Lorentzian expected due to the \textit{T} the development of short-range correlations between the \textit{g} by the relation \chi''(\omega)/\omega \propto C_m T I_{q}(\omega), where I_{q}(\omega) is the Lorentzian spectral function (see the Methods for details)\textsuperscript{40–42}. A fit to this equation allows evaluating \textit{Cm}(T) from the integration of \chi''(\omega)/\omega scaled by \textit{T}. In Fig. 3d, the resulting \textit{Cm} versus \textit{T} is plotted. We confirm the two peaks at \textit{T}_N = 0.1 J and \textit{T}^* \sim \textit{J}. The high-\textit{T} peak at \textit{T}^* \sim \textit{J} is somewhat higher than that of the theoretical value of 0.6 J (ref. 9). In addition, the predicted topological transition at \textit{T} \sim 0.005 J is pre-empted by the long-range magnetic order at \textit{T}_N = 0.1 J. We ascribe the discrepancy between experiment and theory to residual interactions, which lift the Raman selection rules of probing the Majorana fermions. In spite of the magnetic order, the persistent two-peak structure in \textit{Cm} suggests that the hyperhoneycomb iridates are in proximity to a Kitaev spin liquid phase.

**Discussion**

Having established that \beta- and \gamma-Li_2IrO_3 have fractionalized fermionic excitations, it is due to compare them with spinon excitations in the well-characterized kagome Heisenberg antiferromagnet \textit{ZnCu}_3(OH)_6\textit{Cl}_2 (refs 3,43). In such a system, geometrical frustration is the key element. Despite disparate sources of fractionalized excitations, a number of key features in the spectral shape and temperature dependence of magnetic scattering, as well as in the Fano (anti)resonance of optical phonons (see the asterisks in Fig. 4) are common to \beta-Li_2IrO_3 and \textit{ZnCu}_3(OH)_6\textit{Cl}_2. Both compounds show a broad continuum with a rounded maximum at low energies, as shown in Fig. 4. In \textit{ZnCu}_3(OH)_6\textit{Cl}_2, the low-energy response decreases linearly down to zero frequency and the magnetic continuum extends up to a high-energy cutoff at 6 J with \textit{J} \approx 16 meV. The former property suggests the formation of a gapless spin liquid and the latter the existence of multiple spinon scattering processes\textsuperscript{43,44}. In a similar manner, the low-energy spectral weight of \beta-Li_2IrO_3 drops to zero with a steeper slope. The similar behaviour observed in the two compounds with different spin and lattice topologies may be due the fact that the bare spinon density of states is modified due to Dzyaloshinskii–Moriya interactions and antisite disorder in

**Figure 3** | **A Fano resonance of the 24 meV phonon mode and magnetic specific heat.** (a) Fit of 24 meV phonon to a Fano profile after subtracting a temperature-dependent magnetic background. The 21 meV phonon on a low-energy side of the Fano resonance is fitted together with a Lorentzian profile. The inset depicts a schematic representation of eigenvector of the 24 meV \textit{A}_2 symmetry mode. The amplitude of the vibrations is represented by the arrow length. Golden balls indicate \textit{Ir} ions and red balls are \textit{O} ions. The \textit{Li} atoms are omitted for simplicity. (b) Temperature dependence of the Fano asymmetry 1/|\lambda| plotted together with the two-fermion form \(1-(1-f(\omega))^{1/2}\) (solid line). (c) The energy \(\omega\) and linewidth \(\Gamma\) as a function of temperature. The solid lines are a fit to an anharmonic phonon model. (d) Temperature dependence of the magnetic specific heat \textit{Cm} derived from the Raman conductivity \(\chi''(\omega)/\omega\).
ZnCu$_3$(OH)$_6$Cl$_2$ and other spin-exchange interactions in $\beta$-Li$_2$IrO$_3$. The resemblance becomes less clear for $\gamma$-Li$_2$IrO$_3$, mainly because the anisotropic Kitaev exchange interactions open a large excitation gap in the low-energy excitations.

Next, we discuss the temperature dependence of the magnetic continuum. Irrespective of the spinon topology and spin-exchange type, the three studied compounds share essentially the same phenomenology. The key feature is the evolution of a spinon continuum into a quasi-elastic response with increasing temperature. This is well characterized by the power-law dependence of $\chi^{\text{dyn}}(T) \sim T^\alpha$. The exponent of $\alpha = 1.85 - 2.64$ is not much different comparing the three compounds$^{43}$. As discussed in Supplementary Note 3, this power-law behaviour is inherent to a long-range entangled spin liquid and is completely different from what is expected for conventional magnets. Despite the distinct spinon band structure, the spinon correlations may be not very different between the 2D kagome and the 3D hyperhoneycomb lattice.

The last remark concerns that $\chi^{\text{dyn}}(T)$ of the 3D hyperhoneycomb materials start to deviate from a power-law behaviour at 220 K. At the respective temperature, the magnetic specific heat shows as a broad peak identified as a thermal crossover from a paramagnet to a Kitaev paramagnet. This anomaly is absent in ZnCu$_3$(OH)$_6$Cl$_2$ with a single type of spinon and thus unique to $\beta$- and $\gamma$-Li$_2$IrO$_3$ having two species of Majorana fermions.

In summary, a Raman scattering study of the 3D honeycomb materials $\beta$- and $\gamma$-Li$_2$IrO$_3$ provides evidence for the presence of Majorana fermionic excitations. A polarization, temperature and composition dependence of a magnetic continuum indicates a distinct topology of spinon bands between $\beta$- and $\gamma$-Li$_2$IrO$_3$. The temperature dependence of an integrated Raman response and the two-peak structure in specific heat demonstrate that a thermal fractionalization of spins brings about fermionic excitations and that the 3D harmonic-honeycomb iridates realize proximate spin liquid at elevated temperatures. These results expand the concept of fractionalized quasiparticles to a 3D Kitaev–Heisenberg spin system.

**Methods**

**Samples.** Single crystals of $\beta$-Li$_2$IrO$_3$ were grown by a flux method. The starting materials Li$_2$CO$_3$, IrO$_2$ and LiCl with ratio 10:1:100 were mixed together and pressed into a pellet. The pellet was placed in a crucible, heated to 1,170 °C for 72 h and slowly cooled down to 900 °C in air. Shriny black crystals with a size of 100 μm were obtained on the surface of the pellet. The phase purity and composition of $\beta$- and $\gamma$-Li$_2$IrO$_3$ were confirmed via powder X-ray diffraction. The bulk magnetic susceptibility is presented in Fig. 2g,h of the main text.

**Raman scattering experiment.** A polarized, resolved Raman spectroscopy was employed to detect spin and phonon excitations of single crystals of $\beta$- and $\gamma$-Li$_2$IrO$_3$. Raman scattering experiments were performed in backscattering geometry with the excitation line λ = 532.1 nm of a Nd:YAG (neodymium-doped yttrium aluminium garnet) solid-state laser. The scattered spectra were collected using a micro-Raman spectrometer (Jobin Yvon LabRam) equipped with a liquid-nitrogen-cooled charge-coupled device. A notch filter and a dielectric edge filter were used to reject Rayleigh scattering to a lower cutoff frequency of 60 cm$^{-1}$. The laser beam was focused to a few-micrometre-diameter spot on the surface of the crystal using a ×50 magnification microscope objective. The samples were mounted onto a liquid-He-cooled continuous flow cryostat, while varying the temperature between 6 and 300 K. All Raman spectra were corrected for heating.

**Analysis of quasi-elastic Raman scattering.** Quasi-elastic light scattering arises from different fluctuations of a four-spin time correlation function or fluctuations of the magnetic energy density. According to Reiter$^{36}$ and Halley$^{41}$, a two-spin process leads to scattering intensity for temperatures above the critical temperature:

$$I(\omega) \propto \int_{-\infty}^{\infty} e^{-i\omega t} dt \langle E(k, t) E(k, 0) \rangle,$$

where $E(k, t)$ is a magnetic energy density given by the Fourier transform of $E(t) = \sum_{\mathbf{r}} j_{\mathbf{r}} \Sigma_{\mathbf{r}} \delta (\mathbf{r} - \mathbf{r})$ with the position of the $i$th spin $\mathbf{r}_i$. Applying the fluctuation–dissipation theorem in the hydrodynamic limit$^{48}$, equation (1) is simplified to

$$I(\omega) \propto \frac{\omega}{1 - e^{-\hbar \omega / k_B T}} C_m T D k^2$$

where $\beta = 1/k_B T$, $C_m$ is the magnetic specific heat and $D$ is the thermal diffusion constant $D = K/C_m$ with the magnetic contribution to the thermal conductivity $K$. Equation (2) can be rewritten in terms of a Raman susceptibility $\chi^{\text{R}}(\omega)$,

$$\frac{\chi^{\text{R}}(\omega)}{\omega} \propto C_m T D k^2$$

This relation is employed to extract the magnetic specific heat from the Raman conductivity in the main text.

**Data availability.** The authors declare that the data supporting the findings of this study are available within the article and its Supplementary Information files.
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