Electron-phonon coupling leads to a wide range of phenomena, from Cooper pairing in metals to Jahn-Teller effect in Mott insulators. The Jahn-Teller (JT) effect, arising from coupling of the orbital degrees of freedom of localized electrons to lattice vibrations (“orbital-lattice coupling”), is a major source driving structural phase transitions. Below the JT structural transition temperature $T_{JT}$, the orbital fluctuations are quenched, and resulting orbital order dictates the spin-exchange coupling, which turns out to be instrumental for understanding the magnetic properties of this compound, including metamagnetic behavior, the origin of magnon gaps, etc. In $J_{eff} = 0$ systems, the pseudo-JT effect leads to spin-nematic transition well above magnetic ordering, which may explain the origin of “orbital order” in Ca$_2$RuO$_4$.

The consequences of the Jahn-Teller (JT) orbital-lattice coupling for magnetism of pseudospin $J_{eff} = 1/2$ and $J_{eff} = 0$ compounds are addressed. In the former case, represented by Sr$_2$IrO$_4$, this coupling generates, through the so-called pseudo-JT effect, orthorhombic deformations of a crystal concomitant with magnetic ordering. The orthorhombicity axis is tied to the magnetization and rotates with it under magnetic field. The theory resolves a number of puzzles in Sr$_2$IrO$_4$ such as the origin of in-plane magnetic anisotropy and magnon gaps, metamagnetic transition, etc. In $J_{eff} = 0$ systems, the pseudo-JT effect leads to spin-nematic transition well above magnetic ordering, which may explain the origin of “orbital order” in Ca$_2$RuO$_4$.

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The difference between $A$ and $B$ levels. The “tetravalent”, i.e., unperturbed wavefunctions $|A_+\rangle = \sin \theta |0, \pm \frac{1}{2}\rangle - \cos \theta |1, \mp \frac{1}{2}\rangle$ and $|B_+\rangle = | \pm 1, \pm \frac{1}{2}\rangle$, in terms of $t_{2g}$ orbital and spin quantum numbers.

Next, we inspect how the shape-distortions of the ground state wavefunction $\tilde{A}$ affect the pseudospin interactions. Deformations are assumed to be quasi-static (adiabatic approximation). Projecting the Kugel-Khomskii type spin-orbital Hamiltonian, Eq. (3.11) of Ref. 3, onto $A$ subspace, we find $\mathcal{H} = \mathcal{H}_s + \mathcal{H}_{s-1}$. $\mathcal{H}_s$ comprises the nearest-neighbor Heisenberg $J_z$, Ising $J_x$, Dzyaloshinskii-Moriya $D$, and pseudodipolar $K$ terms

$$J \tilde{S}_i \cdot \tilde{S}_j + J_z S_i^z S_j^z + D \cdot [\tilde{S}_i \times \tilde{S}_j] + K(\tilde{S}_i \cdot \tilde{r}_{ij})(\tilde{S}_j \cdot \tilde{r}_{ij})$$

(3)
derived earlier 18, while

$$\mathcal{H}^{ij}_{s-1} = g_1 \varepsilon_1 (S_i^x S_j^x + S_i^y S_j^y) + g_2 \varepsilon_2 (S_i^x S_j^y - S_i^y S_j^x)$$

(4)
constitutes the (pseudo)spin-lattice interaction that we are looking for. It linearly couples the spin quadrupoles $Q_i^x$ and $Q_i^y$ of $xy$ and $x^2-y^2$ symmetries to corresponding lattice deformations. In essence, $\mathcal{H}_{s-1}$ is nothing but $\mathcal{H}_{JT}$ “reincarnated” as a spin-lattice coupling in $J_{eff} = 1/2$ insulator. The coupling constants $g$ are renormalized from $g$ of Eq. 1 to $\tilde{g} = \kappa g$ by $\kappa = \frac{2}{T} \frac{\sin^2 \theta}{E_{\omega/2}}$ at $t$, $U$, and $J_H$ are hopping amplitude, Coulomb repulsion, and Hund’s coupling, respectively. Roughly, we estimate $\kappa \sim 5 \times 10^{-3}$ and hence $\tilde{g} \sim 25$ meV in Sr$_2$IrO$_4$, using $g \sim 5$ eV typical for $t_{2g}$ systems. In $J_{eff} = 1/2$ compounds based on 4d Ru$^{3+}$ and 3d Co$^{2+}$ ions, $\kappa$ and $\tilde{g}$ should increase as $1/\lambda$.

Breaking tetragonal symmetry. Having derived spin-lattice interaction $\mathcal{H}_{s-1}$, we discuss now its consequences for low-energy properties of Sr$_2$IrO$_4$. First of all, just as the JT coupling, it should lead to the structural instability as soon as the spin quadrupolar moments $Q_i^x$ develop within the (quasi) long-range ordered magnetic domains. Denoting the staggered moment direction by $\alpha$, $\tilde{n} = \langle \cos \alpha, \sin \alpha \rangle$, we find $\langle Q_i^x \rangle = -S^2 \sin 2\alpha$ and $\langle Q_i^y \rangle = -S^2 \cos 2\alpha$ per bond. From Eq. 4 and elastic energy $\frac{1}{2}K_\gamma \varepsilon_1^2$, the spin-lattice induced orthorhombic deformations follow:

$$\langle \varepsilon_1 \rangle = \frac{1}{g_1} \sin 2\alpha, \quad \langle \varepsilon_2 \rangle = \frac{1}{g_2} \cos 2\alpha,$$

(5)where $\Gamma_\gamma = 2S^2g_1^2/K_\gamma$. A mean-field part of $\mathcal{H}_{s-1}$ (4) reads then as follows:

$$\Gamma_1 \sin 2\alpha \left( S_i^x S_j^y + S_i^y S_j^x + \frac{1}{2} \right) + \Gamma_2 \cos 2\alpha \left( S_i^x S_j^y - S_i^y S_j^x \right),$$

(6)with $\alpha$ to be obtained by minimizing the ground state energy $E(\alpha)$. Classically, $E(\alpha) = const + S^2 (\Gamma_1 - \Gamma_2) \cos 2\alpha$. For $\Gamma_1 > \Gamma_2$, $E(\alpha)$ is minimized at $\alpha = 45^\circ$, which is exactly the case of Sr$_2$IrO$_4$ 13, 19. Our theory predicts then $\varepsilon_1$-type ($b > a$) orthorhombic distortion, as depicted in Fig. 1(c). This type of distortion is natural for perovskites, as it does not affect the Me-O-Me bond length.

Breaking $C_4$ symmetry by spin-lattice coupling opens the in-plane magnon gap already on a level of linear spin-wave theory. Eqs. 3 and 6 give $\omega_{ab} \approx 85 \sqrt{\Gamma_1}$. With $\omega_{ab} \sim 2.1 - 2.4$ meV 20, 21 and $\epsilon_{12} \sim 10^{-4}$ 22, we evaluate $\Gamma_1 \sim 3$ meV. Eq. 5 predicts then the spin-lattice induced distortion of the order of $\varepsilon_1 \sim 10^{-4}$ 22. The two-fold $C_2$ anisotropy of magnetoresistivity 23 and the signatures of orthorhombic distortions 27, 28 in Sr$_2$IrO$_4$ find a natural explanation within our theory. Future experiments using, e.g., Larmor diffraction 29 should be able to quantify $\varepsilon_1$ directly. We note also that the deformation induced magnon gap $\omega_{ab}$ far exceeds interlayer couplings 30, and should therefore be essential for establishing the magnetic order at high $T_m \sim 240$ K.

To summarize up to now, the combined action of spin-orbit and JT couplings results in the interaction between magnetic quadrupoles and lattice deformation. Dynamical, coupled oscillations of the $\tilde{n}$-moment direction and lattice vibrations (magnetoacoustic effects 31, 32) are expected; this is an interesting topic for future research. Most importantly, a structural instability is inevitable no matter how large SOC is; this invalidates a common assertion that high tetragonal symmetry of $J_{eff} = 1/2$ system Sr$_2$IrO$_4$ is protected by large SOC.
Metamagnetic transition, in-plane magnon gap.— We discuss now further manifestations of magnetoelastic coupling in Sr$_2$IrO$_4$. Via spin-lattice coupling, the reorientations of moments under external magnetic field will affect lattice deformations. The latter, in turn, modifies the magnetic anisotropy potential. Such feedback effects result in a non-monotonic behavior of magnetization $M(H)$. In Sr$_2$IrO$_4$, spins are canted by angle $\varphi \simeq D/2J \sim 12^\circ$ [33], see Fig. 2(a,b). Magnetic field couples to the canted moments $\vec{m}$. To calculate $M(H)$, we use a simple model in Fig. 2(b) for the interlayer coupling. The total energy $E$ depends now on two angles $\alpha$ and $\alpha'$, corresponding to the moment directions in different layers, and the field direction $\beta$. We find $E(\alpha, \alpha', \beta)=const + \frac{D}{2} F$, with

$$F = \sin \varphi h_c \cos(\alpha-\alpha') - h \cos(\alpha-\beta) - h \cos(\alpha'-\beta) - \frac{D}{2} \left[\Gamma_1(\sin 2\alpha + \sin 2\alpha')^2 + \Gamma_2(\cos 2\alpha + \cos 2\alpha')^2\right].$$

Here, $h = g\mu_B H$, and $h_c = 4J_c S \sin \varphi$ is the interlayer field. Minimization of $F$ gives $\alpha$ and $\alpha'$ as a function of $H$, from which the canted moments $\vec{m}$ and $\vec{m}'$ on different planes and total magnetization $M$ follow. The deformations $\varepsilon_1$ and $\varepsilon_2$ are given by Eq. 8 where $\sin 2\alpha$ and $\cos 2\alpha$ replaced now by $\frac{1}{2}(\sin 2\alpha + \sin 2\alpha')$ and $\frac{1}{2}(\cos 2\alpha + \cos 2\alpha')$, respectively; this implies the field-dependence of the deformations (magnetostriction).

Fig. 2(c) shows $M(H)/M_0$ calculated with $h_c = 18 \mu$eV ($\approx 0.16$ T). Without spin-lattice coupling, $\vec{m}$ and $\vec{m}'$ gradually rotate towards each other and $M$ grows monotonically. Spin-lattice induced anisotropy results in a metamagnetic transition as observed [15, 19]. At $H = H_{cr}$, $\vec{m}$ and $\vec{m}'$ flip and become parallel. For $\Gamma_1 > \Gamma_2$ as in Sr$_2$IrO$_4$, $H_{cr}$ for easy-axis $b$ is lower than that for hard-axis; this result has recently been confirmed experimentally [34]. We note that $M(H)$ near $H_{cr}$ is sensitive to angle $\beta$, so the quenched disorder and sample alignment issues should be relevant in the data analysis.

Next, we discuss the in-plane magnon gaps generated by spin-lattice coupling $H_{an-1}$. Due to interlayer coupling, there are two different modes. At small fields, $H \ll H_{cr}$, the optical and acoustic mode gaps are $8S \sqrt{J(\Gamma_1 + \frac{\sin \varphi}{4S} h_c)}$ and $8S \sqrt{\Gamma_2}$, respectively. Above the metamagnetic transition, $H \geq H_{cr}$, we find

$$\omega_{ab}^\pm \simeq 8S \sqrt{J(\Gamma(\alpha) + \frac{\sin \varphi}{4S}[h \cos(\alpha-\beta) - h_c \pm h_c])}. \quad (8)$$

Here, $\Gamma(\alpha) = \Gamma_1 \sin^2 2\alpha + \Gamma_2 \cos^2 2\alpha$, and $\alpha$ follows from $2S(\Gamma_1 - \Gamma_2) \sin 4\alpha = h \sin \varphi \sin(\alpha-\beta)$. For $H \parallel b$, this gives $\alpha = \beta (-\frac{\pi}{2})$ and $\Gamma(\alpha) = \Gamma_1$. For $H$ along $y$ axis, $\alpha \sim \beta = 0$, and $\Gamma(\alpha) \sim \Gamma_2$; this implies weak distortion $\varepsilon_2$ and smaller magnon gap. The main message is that the magnon gaps become strongly dependent on the field direction, as shown in Fig. 3. The above equations should help to quantify $\Gamma_1$ and $\Gamma_2$ from experiments. The results in Fig. 3 are qualitatively consistent with the recent Raman data [20], a detailed analysis would require a derivation of the Raman matrix elements necessary for the mode assignment.

Via the magnetoelastic coupling, quasi-2D AF correlations above $T_m$ [22, 35] should lead to slowly fluctuating lattice deformations (see Fig. 1), which, in turn, will affect phonon dynamics. Indeed, strong Fano anomalies of phonons have been observed in Sr$_2$IrO$_4$ [39].
Spin-nematic order in \( J_{\text{eff}} = 0 \) systems.—Finally, we move to pseudospin \( J_{\text{eff}} = 0 \) case, and show that, despite having neither orbital nor spin degeneracy, the JT coupling is relevant even here. In general, the \( J_{\text{eff}} = 0 \) compounds are of interest because they host "excitonic" magnetism \[37\] - magnetic order via condensation of spin-orbit \( J_{\text{eff}} = 0 \rightarrow 1 \) excitations. The expected non-Heisenberg type magnon and amplitude (Higgs) modes have been observed in Ca\(_2\)RuO\(_4\) \[38, 39\]. Also, \( J_{\text{eff}} = 0 \) systems illustrate well the interplay between three "grand forces" in Mott insulators - the JT coupling, spin-orbital exchange interaction, and spin-orbit coupling \[3\].

As a toy model, we consider 2D square lattice of \( J_{\text{eff}} = 0 \) ions (e.g., \( d^3 \) Ru\(^{4+}\)) in an octahedral field. The \( t_{2g} \) orbital configuration is subject to the JT effect; however, it is opposed by SOC that favors spin-orbit singlet \( J_{\text{eff}} = 0 \) instead \[37, 40\]. This competition can be resolved by mixing the \( J_{\text{eff}} = 0 \) wavefunction with the excited \( J_{\text{eff}} = 1 \) states, by virtue of spin-orbital exchange interactions. Since \( J_{\text{eff}} = 1 \) level hosts a quadrupolar moment, the ground state becomes JT active, and the moment (quantified by color intensity) and XY-type magnon.

Regard the JT coupling, we consider a tetragonal distortion \( \varepsilon = \frac{a+b-2c}{a+b+c} \), which splits the \( xy \) and \( xz/yz \) orbital levels by \( g_{\varepsilon} \): \( H_{\text{JT}} = g_{\varepsilon} \frac{1}{2} (n_{xx} + n_{yy} - 2n_{xy}) \). In the spin-orbit entangled \( T \)-basis, this coupling lowers \( J_{\text{eff}} = 0 \) singlet by \( E_s = (\frac{J}{2} - \frac{\lambda}{2} - \sqrt{\frac{J}{2} - \frac{\lambda}{2} + \delta + \delta^2}) \lambda \), and splits \( J_{\text{eff}} = 1 \) triplet into \( T_{x/y} \) doublet and \( T_z \) singlet by \( \Delta_z = (\delta + \sqrt{1 + \delta^2} - 1) \lambda \), where \( \delta = g_{\varepsilon}J/2 \). As a result, the spin gap reduces from \( J \) to \( E = (\frac{1}{2} + \frac{\lambda}{2} - \delta + \delta^2 - \sqrt{1 + \delta^2}) \lambda \), see Fig. 4(a). At the critical value of \( E \sim J \), the \( T_{x/y} \) doublet condenses, forming a ground state with finite quadrupole moment \( Q = n_{x/z}^2 + n_{y}^2 - 2n_{xy}^2 \). While the cubic symmetry may be broken at finite temperature \( T_{\text{JT}} \), long-range magnetic order is delayed due to XY-type phase fluctuations; therefore, \( T_m \) and \( T_{\text{JT}} \) are separated in quasi-2D \( J_{\text{eff}} = 0 \) systems. We think that the "orbital order" in Ca\(_2\)RuO\(_4\) near 260 K \[42\], well above \( T_m \), is in fact the JT driven spin-nematic order. The observed XY-type magnons \[38\] further support the picture of spin-orbit entangled \( T_{x/y} \) condensate.

A mean-field phase diagram of \( H_{\lambda,J} + H_{\text{JT}} \), supplemented by the elastic energy \( \frac{1}{2} K_x^2 \), is shown in Fig. 4(b) as a function of \( J/\lambda \) and \( E_{\text{JT}}/\lambda \). \( E_{\text{JT}} = \Delta/3 \) is the JT stabilization energy, where \( \Delta = \frac{2g_{\varepsilon}J}{\lambda} \) is the \( t_{2g} \) orbital splitting at \( \varepsilon = 0 \). At small \( J \) and \( E_{\text{JT}} \), SOC imposes the \( J_{\text{eff}} = 0 \) phase I; at large \( E_{\text{JT}} \), it gives way to the JT-distorted nonmagnetic phase II with finite spin-gap \( E \). In phase III, stabilized by a combined action of the exchange and JT couplings, XY-type magnetic condensate is formed.

Interestingly, the observed magnon bandwidth \( \sim 2J \sim 50 \) meV \[38\] and ratio \( \Delta/2\lambda \sim 2 \) \[38, 41\] locate Ca\(_2\)RuO\(_4\) in the critical area of the phase diagram [see Fig. 4(b)]. This suggests that an unusual magnetism \[38, 39\] and extreme sensitivity of Ca\(_2\)RuO\(_4\) to external perturbations \[45, 46\] are caused by frustration among the JT, spin-orbit, and exchange interactions, further boosted by its proximity to metal-insulator transition.

To conclude, in contrast to the common wisdom, the JT coupling remains an essential part of the low-energy physics in spin-orbit \( J_{\text{eff}} = 1/2 \), and even \( J_{\text{eff}} = 0 \), Mott insulators. Converted into pseudospin-lattice coupling via spin-orbit entanglement, it leads to the structural transitions and magnetoelastic effects. We have shown that the JT coupling resolves hitherto unexplained puzzles of \( J_{\text{eff}} = 1/2 \) Sr\(_2\)IrO\(_4\), and is essential for the phase behavior of \( J_{\text{eff}} = 0 \) Ca\(_2\)RuO\(_4\). This leads us to believe that pseudospin-lattice coupling should be generic to a broad class of spin-orbit \( J_{\text{eff}} \) compounds, including the Kitaev-model materials of high current interest \[47, 48\].

In the latter, the pseudospins are highly frustrated, and their coupling to lattice lead to more radical effects than in conventional, unfrustrated magnets like Sr\(_2\)IrO\(_4\).
We thank B. J. Kim, J. Porras, J. Bertinshaw, B. Keimer, J. Chaloupka, and O. P. Sushkov for discussions. We acknowledge support by the European Research Council under Advanced Grant No. 669550 (Com4Com).

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