Frustrated magnets, in which classical ground states have multiple accidental degeneracies, have been at the focus or renewed attention. While a large body of theoretical work has focused on the lifting of this degeneracy by thermal or quantum fluctuations ('order by disorder') in real materials, alternate mechanisms might dominate. One mechanism that is always present is the coupling of magnetism to the lattice (spin-phonon coupling). Exploring the ramifications of such couplings is still in its infancy and leads to new theoretical problems with consequences for real materials. Finally, the subject is potentially important for future applications - spin phonon couplings could give rise to multiferroic systems, where magnetic order is coupled to ferroelectricity.

The role of lattice distortions in promoting valence bond physics (spin Pierels effect) is well known and consequences for spin 1/2 frustrated quantum magnets have been studied. In the opposite large spin (semi-classical) limit, the role of lattice distortions in selecting the colinear ground states amongst the vast number of degenerate spin configurations of the pyrochlore magnet has been emphasized. Further selection of a unique colinear ground by various spin phonon interactions have also been proposed.

Here we will study the effect of spin-phonon interactions on the classical ground states of the triangular and Kagome magnets. In contrast to earlier applications, all ground states of the antiferromagnets on these lattices are non-colinear (120°) configurations. Hence a finite strength of spin phonon coupling is required to induce colinear states. We believe this to be the origin of the puzzling colinear magnetism seen in several triangular lattice materials, such as CuFeO$_2$, $\alpha$-NaFeO$_2$, and MnBr$_2$. In these materials with $S = 5/2$ filled shell moments, magnetic anisotropy is likely to be (and in some cases known to be) very small, and cannot be invoked to explain the colinear order. Furthermore, we study the precise pattern of the colinear magnetic order induced within the Einstein Site Phonon (ESP) model, which builds in an energy cost for displacing a magnetic atom from its ideal lattice position, and is parameterized by a single coupling constant. Analytic arguments combined with numerical calculations (simulated annealing) are used to establish the zero temperature phase diagram of the classical spin models - which is briefly summarized below. On the triangular lattice, increasing the spin-phonon coupling leads to the phase diagram in Figure 1, where the four sublattice zig-zag state (Z state) is the first colinear state selected. At large couplings a four sublattice parallel stripe state (S state) of up and down spins is obtained. The phase diagram in a magnetic field is remarkably complex, with magnetization plateaus at 1/5th, 1/3rd, 3/7th, 3/5th, 5/7th, and 1/2 occur in the parameter range where the Z state appears, and also 1/9th and 7/15 at larger spin-phonon couplings. In particular, the four sublattice Z state is typically converted into a 1/5th plateau with a five sublattice spin structure on increasing the field.

These are precisely the two colinear states observed in CuFeO$_2$, an extensively studied triangular magnet, where the complex four and five sublattice colinear structures described above was seen. This was previously rather mysterious since the only available models which captured such orders were Ising Hamiltonians with large and very specific second and third neighbor interactions ($J_2 = 0.45J_1$, $J_3 = 0.75J_1$) (or more complicated models, see e.g. Plumer) which are unnatural since the magnetic susceptibility is nearly isotropic in the paramagnetic state. In contrast, our isotropic spin phon model involves a single parameter - the spin phonon coupling. Predictions for higher magnetization plateaus and other states for this material are made. The magnetically induced electrical polarization observed in this system is however not captured by our simple model, pointing to the role of other interactions which we briefly discuss. The $S$ state is precisely the spin arrangement seen at low temperatures in $\alpha$–NaFeO$_2$ and MnBr$_2$. Predictions for magnetic plateaus in these materials are also made.
On the Kagome lattice too we find that beyond a critical coupling, colinear ground states are obtained, but in contrast to the triangular lattice, the manifold of these states has an extensive entropy. Only at larger couplings is there a transition into a unique ground state. This may be of relevance to the recently studied Kagome staircase compound, Mn$_3$V$_2$O$_6$ [13]. If the spin phonon mechanism is as widely relevant as some of our results suggest, theoretical studies will have to move beyond their current focus on purely rigid lattice models.

**Spin Phonon Model:** The minimal spin phonon coupling arises from the dependence of the exchange coupling on separation between the magnetic ions $\vec{J}(r)$. If sites $i$ and $j$ approach each other by $u_{ij}$, then the spin phonon Hamiltonian then is:

$$H = \vec{J} \sum_{<ij>} (1 - \alpha_{ij} u_{ij}) \vec{S}_i \cdot \vec{S}_j + H_{lattice}(\{\vec{u}_i\})$$  \hspace{1cm} (1)

where $\alpha = -J^{-1} \partial J/\partial r$, $H_{lattice}$ is the phonon Hamiltonian and $\vec{u}_i$ is the displacement of site $i$. Hence, $u_{ij} = (\vec{u}_i - \vec{u}_j) \cdot \hat{e}_{ij}$ is the relative displacement projected on the bond $ij$ ($\hat{e}_{ij}$ is the unit vector from site $i$ to $j$). Two types of phonon Hamiltonians have been proposed. First, the bond phonon model of Penc et al. [3] where $H_{lattice} = (K/2) \sum_{<ij>} u_{ij}^2$. This model assumes that bond displacements are independent, which may be an oversimplification for many lattices. Integrating out the phonon degree of freedom here generates just the bi-quadratic term $-bJ \sum_{<ij>} (\vec{S}_i \cdot \vec{S}_j)^2$. (The phonon dynamics can be neglected if its frequency is much larger than magnetic energy scales). While for sufficiently large $b$, colinear states result, the model is still highly degenerate (Ising antiferromagnet ground states on the triangular or Kagome magnets). The selection by quantum fluctuations will be discussed elsewhere [14]. Instead, here we turn to the second model, the ESP model of Bergman et al. [7]

$$H_{lattice} = (K/2) \sum_i u_i^2$$  \hspace{1cm} (2)

which in contrast to the bond phonon model, respects the inevitable correlations between bond length arising from the underlying lattice structure. The ESP model describes a dispersionless optical phonon branch. More realistic phonon model informed by the detailed crystal structure of the material and acoustic phonons could generate longer-range effective spin interactions that are hard to deal with. In the interests of simplicity and generality we will restrict attention to the ESP model. Integrating out the lattice displacement $\vec{u}_i$ results in the effective spin Hamiltonian:

$$H_{ESP} = \vec{J} \left[ \sum_{<ij>} \vec{S}_i \cdot \vec{S}_j - \tilde{c} \sum_i \vec{F}_i^2 \right]$$  \hspace{1cm} (3)

where $\tilde{c} = \alpha^2 J/(2K)$ is a positive dimensionless coupling and we have defined the dimensionless ‘force’ on site $i$ as $\vec{F}_i = \sum_{j \in \text{neighbors of } i} (\vec{S}_i \cdot \vec{S}_j) \hat{e}_{ij}$, the sum in this definition is over the (six) neighbors of site $i$. The spin-phonon interaction seeks to maximize the force $\vec{F}$, which will result in gaining the maximum spin interaction energy. Note, the second term in Eqn. 3 generates interactions involving three adjacent spins $\hat{e}_{ij} \cdot \hat{e}_{jk} (\vec{S}_i \cdot \vec{S}_j)(\vec{S}_j \cdot \vec{S}_k)$.

**Triangular Lattice:** In the following we will consider a single triangular lattice sheet governed by the Hamiltonian in Eqn. 3 i.e. with nearest neighbor antiferromagnetic interactions and additional interactions generated by the spin-phonon term. We focus on the zero temperature phase diagram, as a function of the single parameter $c$ and subsequently in an applied magnetic field. Since we are primarily interested in large spin (eg. $S = \frac{3}{2}$), we focus on classical spins, where we can write $\vec{S}_i = S \hat{n}_i$, where $\hat{n}_i$ is a unit vector. This leads to an extended classical Heisenberg model of unit vectors with rescaled couplings $J = S^2 J$ and $c = S^2 \tilde{c}$. If we choose to restrict to Ising states, $\vec{S}_i = \sigma_i \hat{S}_i$, this effective spin Hamiltonian simplifies to an Ising model with nearest-, second- and third-neighbor coupling, $J - cJ$, $cJ$ and $cJ$ respectively. However, in this model the second and third neighbor couplings are constrained to be strictly equal. This might be a rationalization for the large second- and third-neighbor couplings used in previous Ising models [11].

Consider the ground state for classical spins on raising $c$. While at $c = 0$ the the regular 120° pattern of O(3) spins on the triangular lattice is realized, this is expected to survive to finite $c$ as well. The ground state energy per site is $E_0/J = -3/2$, and since the force vanishes in this state, it is independent of $c$. While a full numerical solution is required (and provided below) for the phase diagram of this model, we can make some plausible analytic arguments which will be confirmed by the numerics. Clearly, colinear states are preferred for large $c$ since they give rise to the maximum force. However, near the phase boundary with the 120° states, the exchange $J$ will presumably be important, and hence we restrict attention to those colinear states that best satisfy $J$. These are nothing but the ground states of the triangular lattice Ising antiferromagnet (TLIA) with nearest neighbor exchange, with two (one) up and one (two) down spins per triangle. We can then ask, which configuration within this manifold optimizes the force term? This question can be rigorously answered - it is the Z state. Using the dimer representation of the TLIA states, where a dimer is drawn orthogonal to each unsatisfied bond, and leads to a hard core dimer configuration on the honeycomb lattice, we see that the ‘force’ on site $i$ is determined by the dimer configurations on the hexagon surrounding site $i$. By checking the possible dimer configurations on a hexagon, we see that the force $|\vec{F}_i| = 2$ if there is
one dimer in the hexagon or two not-opposite dimers; otherwise $|\vec{F}_i| = 0$. The ground state maximizes $\sum_i F_i^2$. Hence it must have two dimers in every hexagon (since on average there are two dimers per hexagon and a configuration with one dimer hexagon implies also a hexagon with three dimers, which experiences no force). Combining this with the condition that the two dimers cannot be on opposite sides leads us to the unique zigzag state $Z$, with a four sublattice unit cell as shown in Fig. 1 and ground state energy per site $E_0/J = -1 - 4c$.

The full phase diagram is obtained using simulated annealing on lattices with periodic boundary condition and various sizes up to $10 \times 10$, and choosing the state with lowest energy per site. Simulations on each size were done by an exponential annealing schedule from $\beta J = 0.1$ with a random initial state to $\beta J = 1000$, with a total of $20000 \sim 40000$ sweeps, the whole process was repeated 10 times to ensure stability of results. Site-update with Metropolis dynamics was used. While the algorithm does not guarantee convergence to the ground state we nevertheless believe an accurate picture emerges since all analytic expectations have been met, and we have not been able to guess ground states with better energies.

The numerically obtained phase diagram in zero field is shown in Fig. 1. At $c = 1/8$ the $120^\circ$ state is replaced by the four sublattice $Z$ state, with alternating zig-zags of up and down spins and ground state energy per spin $E_0/J = -1 - 4c$. This is stable till $c = 1/6$, where an 8 sublattice state ($E_0/J = -10c$) takes over. Beyond $c = 1/2$, the $S$ state of up-up-down-down stripes ($E_0/J = 1 - 12c$) is found and persists to large couplings. At the transition points $c=1/6$ and $c=1/8$, there are additional accidental degeneracies, and quantum effects could be important in resolving these.

The phase diagram in a magnetic field $H = -J \sum_i h n_i^z$ is remarkably rich. For small $c$ where the $120^\circ$ state is realized, a highly degenerate set of states well known for the Heisenberg triangular antiferromagnet are obtained. Since they all have vanishing force contributions, the spin phonon interaction does not split this degeneracy. At larger values of $c$, the simulation shows a plethora of plateau states, which we briefly discuss here and leave details to [15]. Interestingly the 1/5-plateau with the pattern observed in CuFeO$_2$ occur for a wide range of parameter $c$. For the parameter interval $0.14 < c < 1/6$ our model shows both the zigzag $Z$ ground state at zero field and the 1/5-plateau in magnetic field, as in CuFeO$_2$. Other prominent plateaus that occur in the range of $c$ where the $Z$ state appears are the 3/7th and 5/7th states with 7 site unit cell, a 1/2 magnetization plateau with an 8 site unit cell and two distinct 3/5th plateaus with 5 sites per unit cell. There is also a small region of 1/3 plateau, with a 12 site unit cell. The evolution of a plateaus state with increasing field can proceed in two ways. Either it can jump directly to another plateau, or undergo a canting transition, where the field direction is not parallel to the staggered moment. For example, the zero field $Z$ state in the fully isotropic model undergoes a spin-flop transition immediately on applying a field, and the staggered moment is orthogonal to the field and the induced uniform magnetization. The evolution of the 1/5 plateau state on increasing the field however is continuous, with a gradual tilting of the staggered component away from the field. This phase boundary can be calculated analytically and agrees very
well with the simulations. Such canted states are labeled \( c - f \) in the figure (where \( f \) refers to the plateau they derive from). Such states are of course absent in Ising model studies \[\text{[11]}\]. Other plateaus occur for larger \( c \), which will be discussed in detail elsewhere \[\text{[15]}\]. Here we note that the 1/9th plateau that extends all the way down to zero field occurs because of the accidental degeneracy at the point \( c = 1/2 \) which includes finite magnetization configurations, with a maximum magnetization of 1/9th, which is selected by the field. Amusingly, the most obvious 1/3 plateau expected for a triangular lattice, consisting of up, up and down spins on the three sublattices, does not occur (the 1/3 plateau at high fields involves a 12 spins).

**CuFeO\(_2\) and other materials:** In CuFeO\(_2\) the 4 sublattice \( Z \) state is observed, which persists in a field up to \( B < 6 \) Tesla. At higher fields \( B > 14 \) Tesla, the 5 sublattice 1/5th magnetization plateau is observed. We note that both these states occur in our spin phonon model when \( 0.14 < c < 1/6 \). To estimate the spin phonon coupling \( c = \alpha^2 J/2K \) in CuFeO\(_2\), we use \( J = 39 \) Kelvin (from the measured Weiss constant \[\text{[17]}\]) and estimate \( \alpha \sim 7 \) and \( K \sim 10^{6} \) Kelvin \[\text{[4]}\], which gives \( c \sim 0.1 \) which is in the right ball park. While an isotropic spin model with magnetic order cannot have a magnetization plateau centered at zero field, even a very small magnetic anisotropy (e.g. an easy axis anisotropy \( -D \sum_i S_i^2 \)) can produce the observed zero magnetization plateau, since the plateau width \( \Delta B \) is readily seen to be \( \Delta B \propto \sqrt{DJ} \). A 1\% anisotropy \( D/J \) produces the right plateau width \[\text{[17]}\]. The magnetization profile as a function of field at \( c = 0.15 \) with a 1\% easy axis anisotropy is shown in Fig. 3 (the field scale \( J/\gamma g_0 S \) is \( \sim 10 \) Tesla per unit from the estimated value of \( J \)). The higher field magnetization plateaus and structures are predicted for future experiments on single crystals. Existing pulsed field measurements on powder samples reveal a sequence of anomalies at different fields up to full polarization, but the magnetization plateaus and structures remain to be conclusively identified \[\text{[15]}\]. Lastly we note that the ferroelectric phase with incommensurate spiral order observed experimentally in CuFeO\(_2\) in the field range \( 7 < B < 14 \) Tesla \[\text{[19]}\] is not produced here, indicating the importance of other lattice couplings e.g. to the oxygen atoms mediating the superexchange interaction. The up up down down stripe pattern for \( 1/2 < c \) has been observed as the ground state for some materials with triangular lattice structure such as \( \alpha\)-NaFeO\(_2\) \[\text{[8]}\] and MnBr\(_2\) \[\text{[10]}\]. Finding the predicted 1/9th and 1/3 plateaus in these materials would be a check of the spin phonon origin of these states.

**Kagome Lattice:** The Einstein site phonon model on the Kagome is virtually identical to the triangular case, except that the lower symmetry in this case (lack of site centered 60° rotation symmetry) allows for anisotropic confining potential on the atoms. For simplicity, we assume an isotropic confining potential, but the main results are independent of this assumption.

Simulated annealing was applied to this model with similar settings as the triangular case. The zero-field phase diagram is presented in Fig. 4. For small \( c \) we still get the ground states of the pure Heisenberg model, which are known to be extensively degenerate. For \( c > 1/12 \) we get colinear states, but in the range \( 1/12 < c < 1/6 \) an extensive degeneracy remains \[\text{[20]}\]. Even more interestingly the zero-field ground states can have arbitrary magnetization ranging from \( -1/9 \) to \( 1/9 \) per site. Therefore, in this zero temperature classical model, applying a small field will immediately induce a 1/9-magnetization-plateau state. We expect that thermal and/or quantum fluctuation can lift this accidental degeneracy which is left for future work \[\text{[15]}\]. Further increasing \( c \) beyond 1/6 pushes the system into a unique colinear states (see Fig. 4).

We have shown that spin lattice couplings can induce a rich variety of complex colinear orders on the triangular and Kagome lattices. The moderate spin-phonon couplings strengths required make this a viable mechanism to explain similar ordering patterns seen in materials like CuFeO\(_2\). Our results underline the need to go beyond the
current focus on purely rigid lattice spin models. Future work will study the effect of quantum fluctuations and valence bond states that naturally arise from disordering these colinear configurations. Including other atomic displacements might account for interesting magnetoelectric phenomena, and guide the search for multiferroic materials. We acknowledge support from LBNL DOE-504108 and useful discussions with K. Damle and R. Cava.

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