Kitaev interaction and possible spin liquid state in CoI_2 and $Co_{2/3}Mg_{1/3}I_2$

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ABSTRACT

It is still a great challenge to explore quantum spin liquid (QSL) state. Here we demonstrate that the van der Waals triangular antiferromagnet CoI_2 hosts a remarkable Kitaev interaction. We find the Co^{2+} ion is in the high spin and $J_{\text{eff}} = 1/2$ state. The ligand mediated t_{2g}-e_g hopping gives rise to a strong Kitaev interaction. This bond-dependent frustration together with the geometric frustration determines the experimental helical AF state of CoI_2. We then propose to suppress the geometric frustration via a partial Mg substitution for Co, and indeed we find that $Co_{2/3}Mg_{1/3}I_2$ has much weakened geometric frustration but the robust bond dependent frustration. As a result, the overwhelming Kitaev interaction renders $Co_{2/3}Mg_{1/3}I_2$ closest to QSL among several candidate materials.

RESULTS

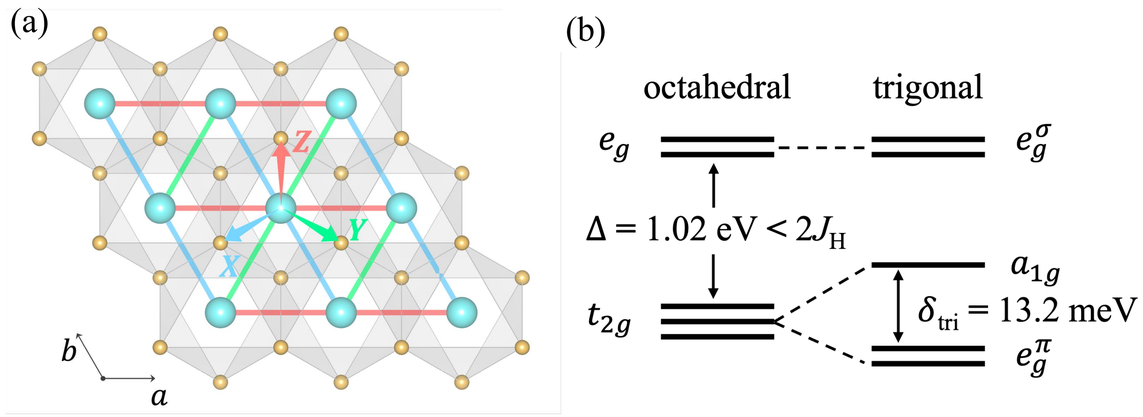

Fig.1. (a) The c-axis view of crystal structure and local basis of CoI_2. (b) Octahedral and trigonal crystal field splitting suggest $S = 3/2$ ground state and small trigonal distortion.

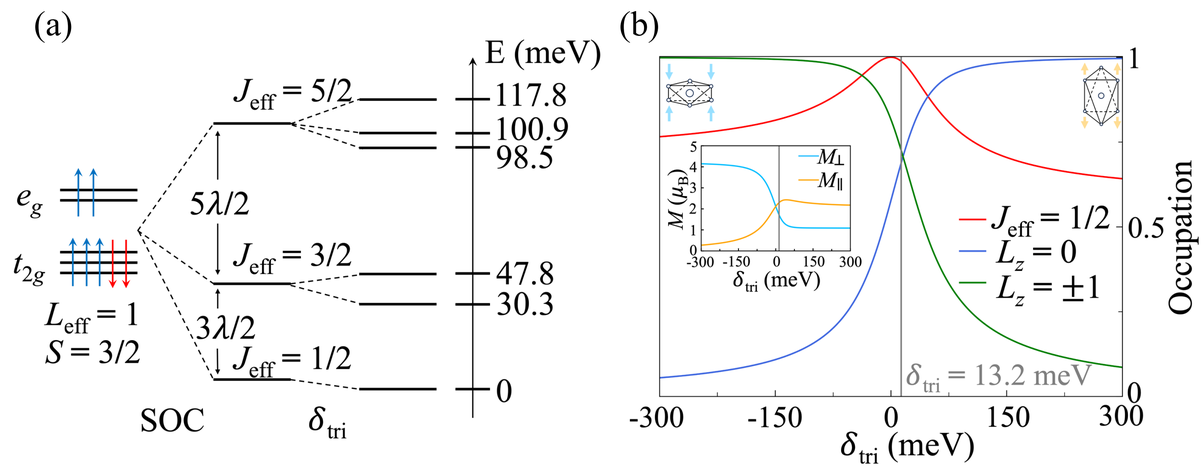

Fig.2. (a) Energy splitting of d^7 high spin state under SOC and trigonal crystal field. (b) The occupation number of ground state under different trigonal crystal field, which indicates $J_{\text{eff}} = 1/2$ ground state of Co^{2+} and weak in-plane anisotropy.

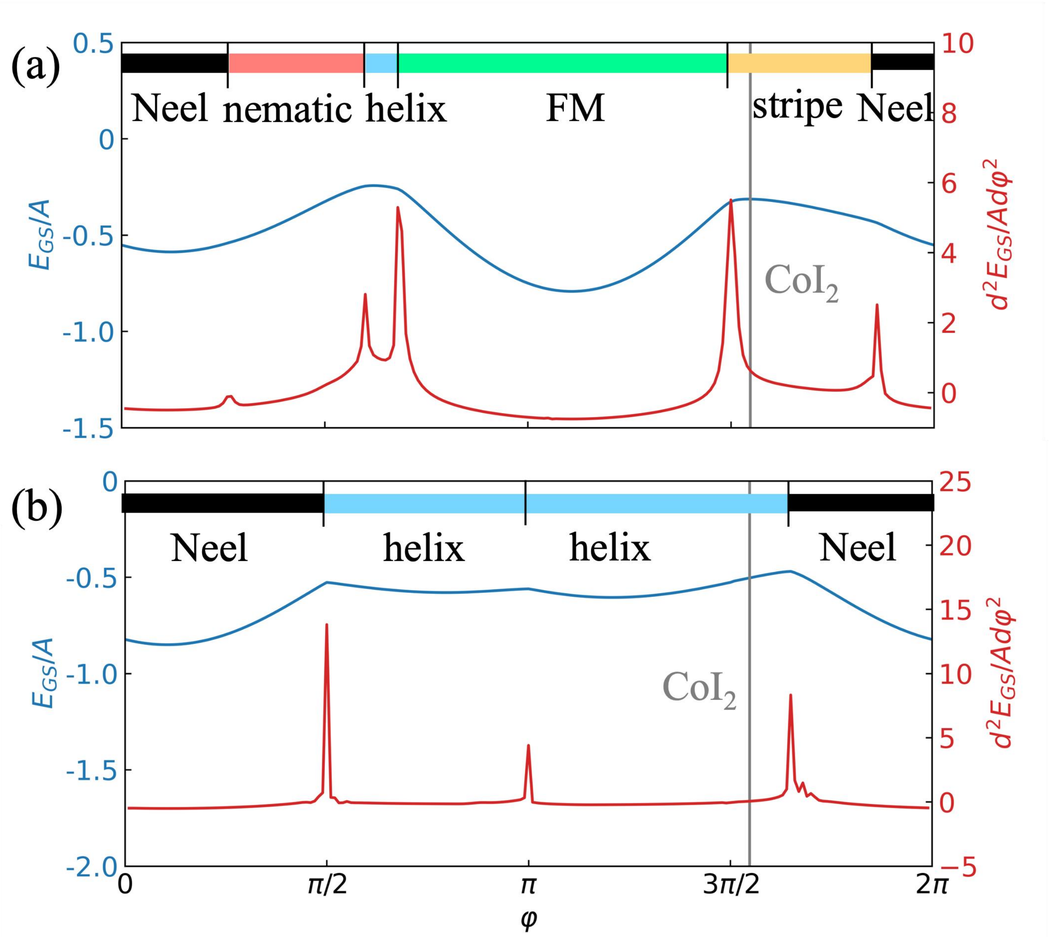

Fig.3. The DMRG phase diagrams of Kitaev-Heisenberg model on triangular lattice (a) without J_3 and (b) with J_3. The competition between Kitaev interaction and J_3 Heisenberg interaction leads to helix magnetic ground state of CoI_2.

	J	K	Γ	Γ'
1NN	0.63 (0.89)	-4.17 (-3.15)	-0.04 (-0.07)	0.06 (0.27)
2NN	-0.15 (-0.21)	-0.56 (-0.32)	0.40 (0.04)	-0.14 (0.02)
3NN	2.16 (0.53)	-0.06 (0.01)	0.38 (0.01)	0.30 (0.06)

Table.1. First, second and third nearest neighbor magnetic interactions in meV for CoI_2 ($Co_{2/3}Mg_{1/3}I_2$). The geometric frustration associated with J_3 interaction is suppressed by Mg doping.

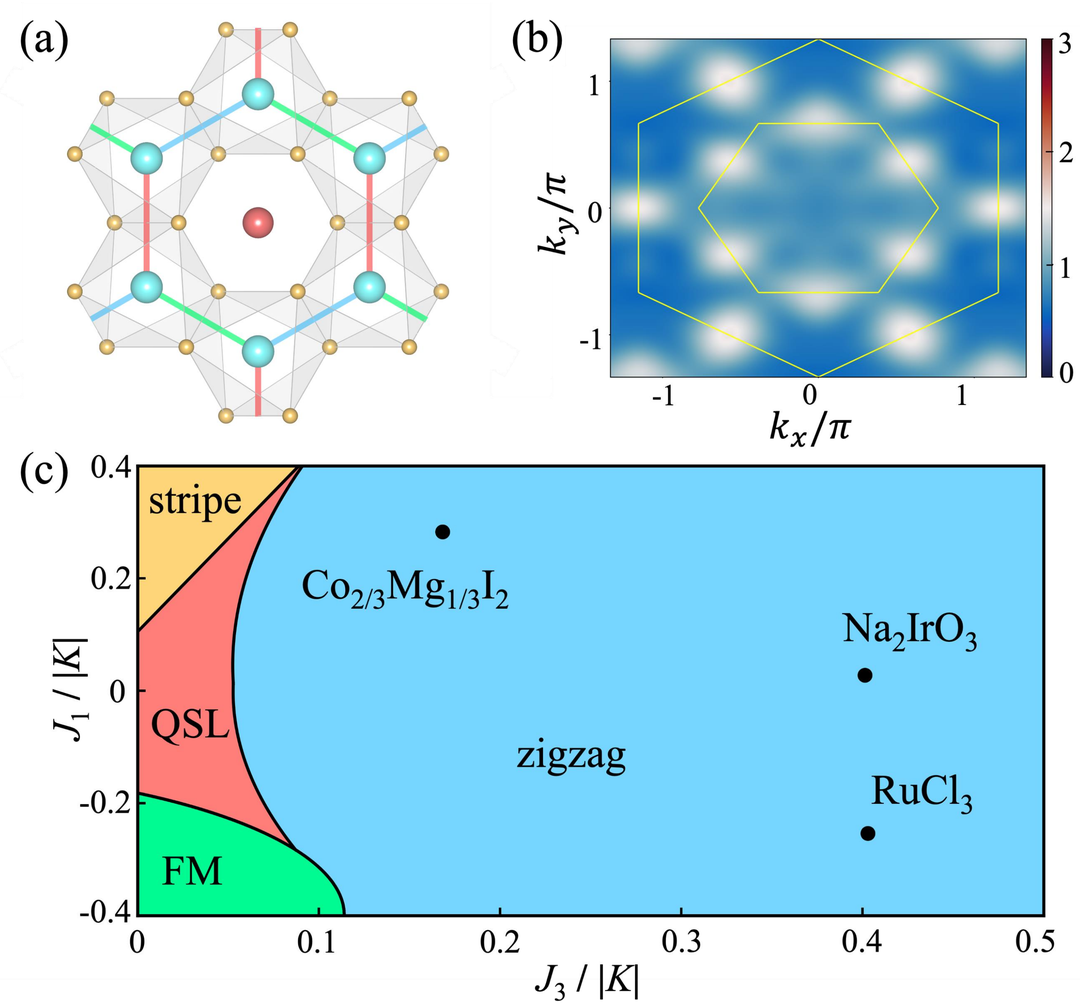

Fig.4. (a) The crystal structure of $Co_{2/3}Mg_{1/3}I_2$. (b) Magnetic structure factor S(q) of $Co_{2/3}Mg_{1/3}I_2$ with 30% J_3. (c) Phase diagram for FM Kitaev interaction K <0 by ED.

CONCLUSIONS

1. CoI_2: strong SOC effect and $J_{\text{eff}} = 1/2$ ground state of Co^{2+}.
2. Kitaev and J_3 interaction leading to helix magnetic structure.
3. $Co_{2/3}Mg_{1/3}I_2$: a better candidate for Kitaev quantum spin liquid.

CoI_2: a new platform for Kitaev quantum spin liquid

REFERENCES

1. Chaebin Kim, *et al.*, Nat. Phys. 19, 1624-1629 (2023).
2. Yaozhenghang Ma[+], Ke Yang[+], Yuxuan Zhou, and Hua Wu*, Phys. Rev. B, 112, L220412 (2025)