Canted magnetization of bulk and monolayer VI_3

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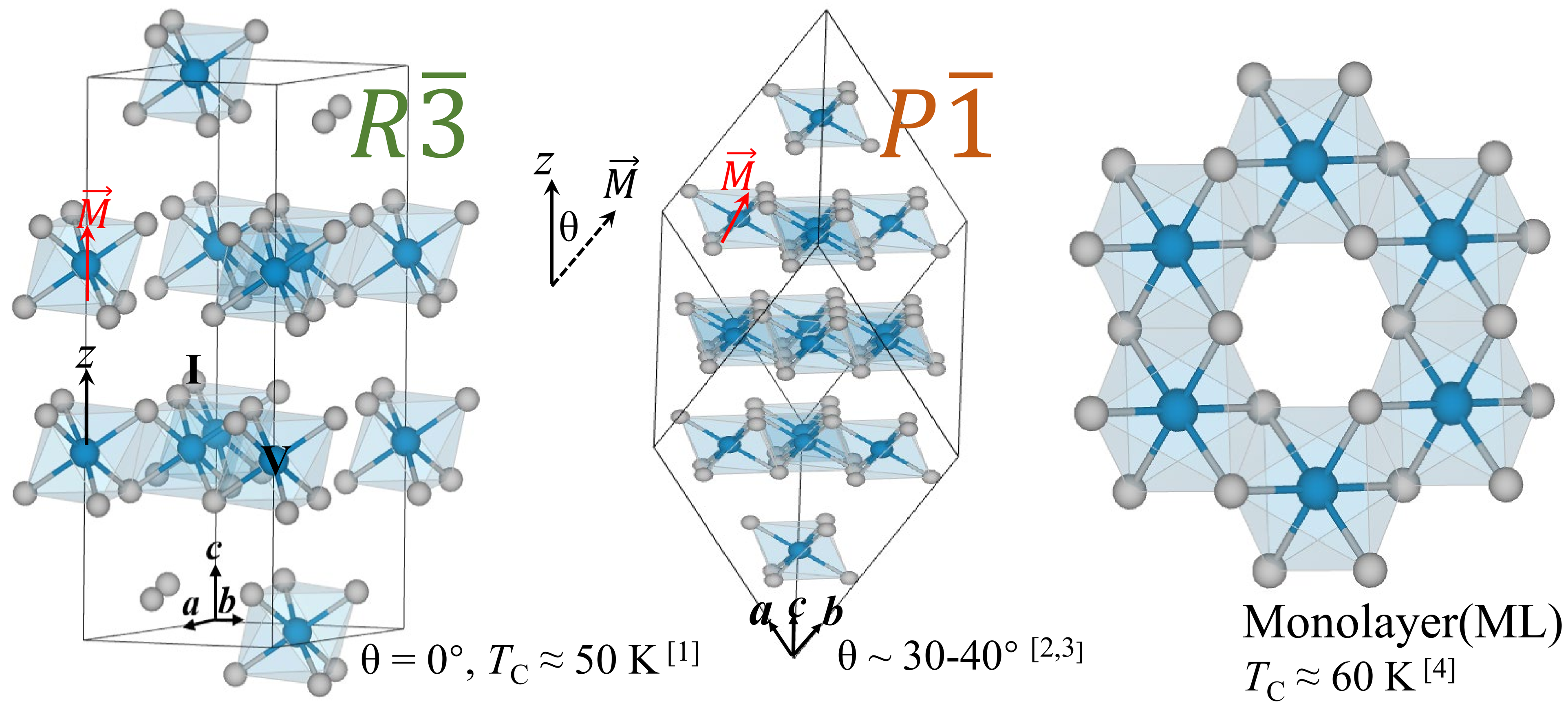
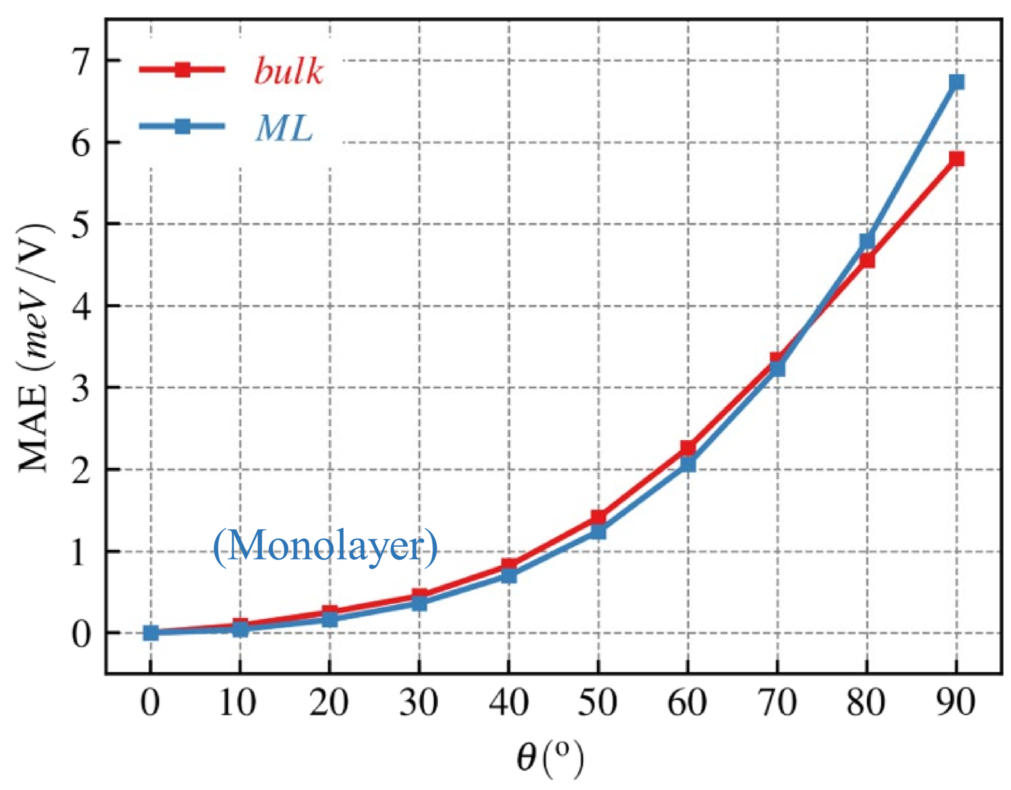
Introduction

Recently, VI_3 has been reported to have a T_C of 50 K in its bulk structure, while its monolayer flake shows a T_C of 60 K, which is higher than that of the bulk phase. In addition, unlike the perpendicular magnetization observed in the $R\bar{3}$ phase, VI_3 with $P\bar{1}$ phase exhibits a canted magnetization. Our first-principle calculations and Hamiltonian analyses confirm a canted magnetization about 30(20)° with z axis for the bulk(monolayer) VI_3 with reduced structural symmetry. The results of renormalized spin wave theory simulations indicate that the large single-ion anisotropy effectively stabilizes the robust T_C in VI_3. We also predict that the anomalous increase of T_C from bulk to monolayer may be attributed to the changes in structure.



DFT Calculations

$R\bar{3}$ phase



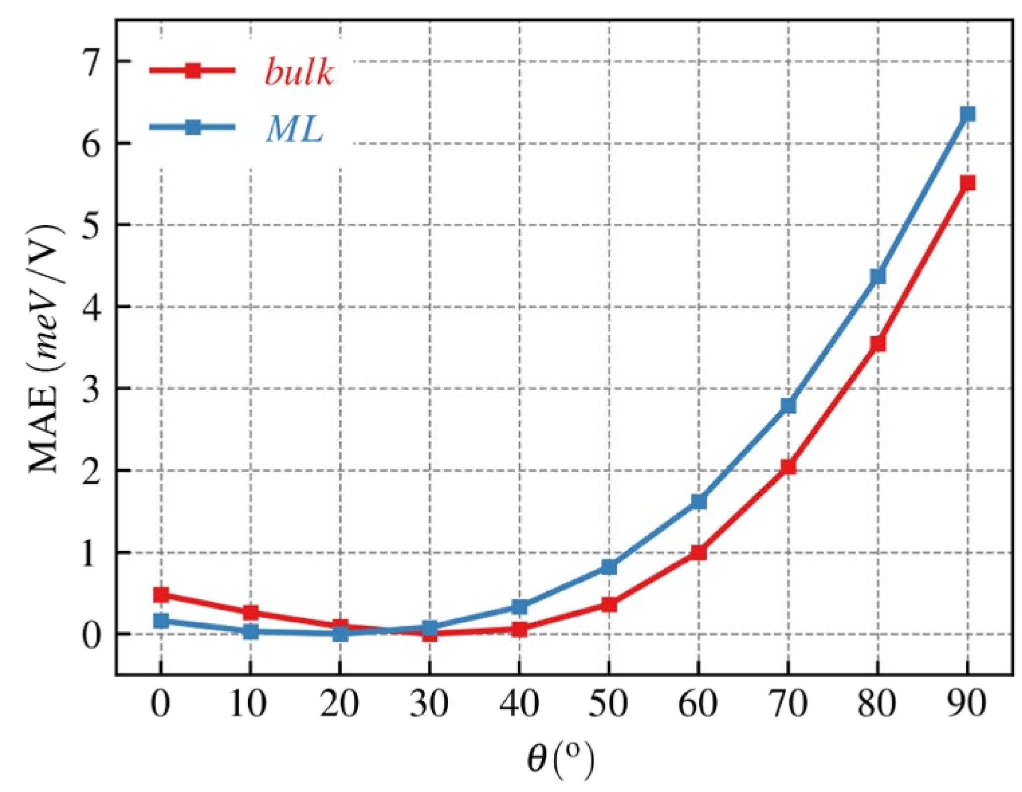
perpendicular $\vec{M} \parallel z$

$a_{1g}^{\uparrow} L_{z-}^{\uparrow}$ ground state

Magnetic anisotropy energy(MAE)

~ 5.80(Bulk) & 6.74(ML) meV/fu

$P\bar{1}$ phase



Bulk:

$\vec{M}$: $\theta \approx 30°$

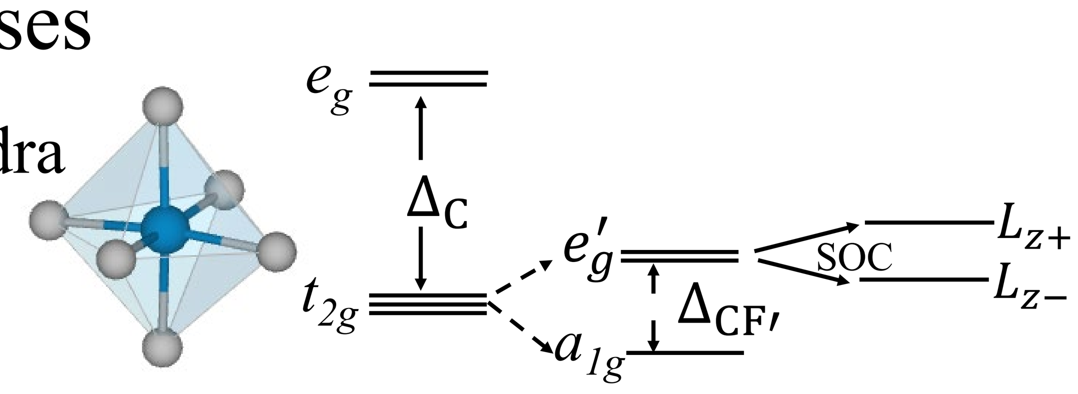
Exp: 36°[2,3]

Monolayer(ML):

$\vec{M}$: $\theta \approx 20°$

Hamiltonian analyses

local octahedra



$R\bar{3}$ phase

H_{tot}	$\|a_{1g}\rangle$	$\|e'_{g1}\rangle$	$\|e'_{g2}\rangle$
$\langle a_{1g}\|$	0	0	$\zeta \cdot i \cdot \sin(\theta)$
$\langle e'_{g1}\|$	0	$\Delta_{CF'}$	$-\zeta \cdot i \cdot \cos(\theta)$
$\langle e'_{g2}\|$	$-\zeta \cdot i \cdot \sin(\theta)$	$\zeta \cdot i \cdot \cos(\theta)$	$\Delta_{CF'}$

Eigenstates:

$|a_{1g}\rangle$

$|L_{z-}\rangle = \frac{1}{\sqrt{2}}(|e'_{g2}\rangle + i|e'_{g1}\rangle)$

$|L_{z+}\rangle = \frac{1}{\sqrt{2}}(|e'_{g2}\rangle - i|e'_{g1}\rangle)$

L_{z+} ——
L_{z-} —↑—
a_{1g} —↑—

$P\bar{1}$ phase

H_{tot}	$\|a_{1g}\rangle$	$\|e'_{g1}\rangle$	$\|e'_{g2}\rangle$
$\langle a_{1g}\|$	0	κ_1	$\kappa_2 + \zeta \cdot i \cdot \sin(\theta)$
$\langle e'_{g1}\|$	κ_1	$\Delta_{CF'}$	$\kappa_3 - \zeta \cdot i \cdot \cos(\theta)$
$\langle e'_{g2}\|$	$\kappa_2 - \zeta \cdot i \cdot \sin(\theta)$	$\kappa_3 + \zeta \cdot i \cdot \cos(\theta)$	$\Delta_{CF'}$

$|L_{z\pm}\rangle = \frac{1}{\sqrt{2}}(|e'_{g2}\rangle \mp i|e'_{g1}\rangle)$

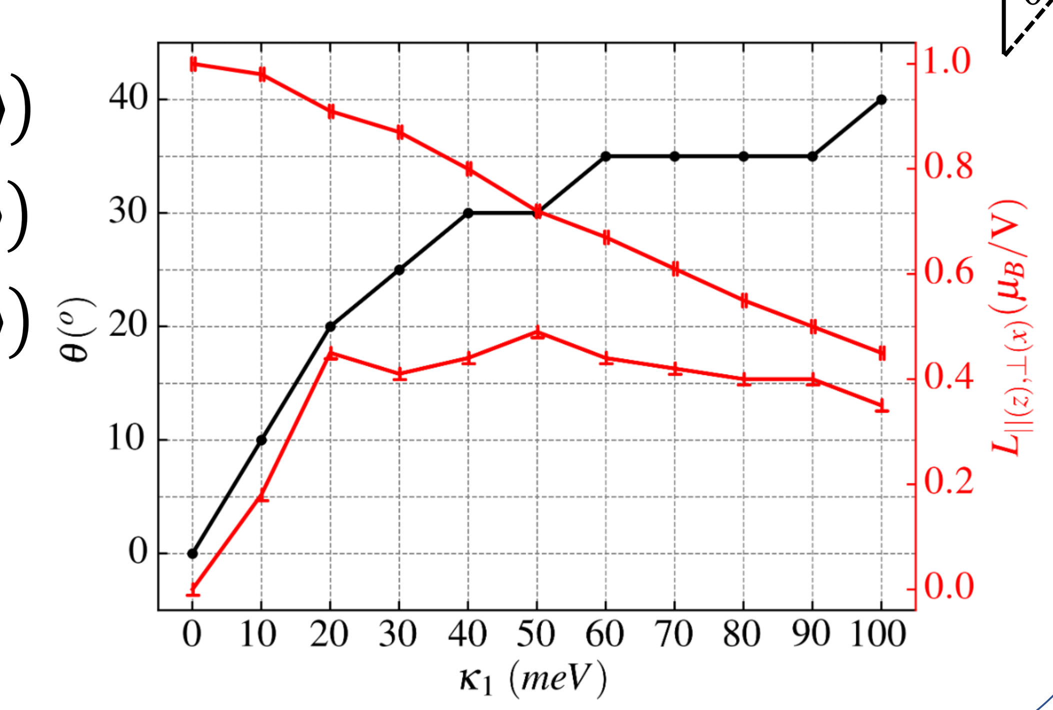
$|L_{x\pm}\rangle = \frac{1}{\sqrt{2}}(|a_{1g}\rangle \mp i|e'_{g2}\rangle)$

$|L_{y\pm}\rangle = \frac{1}{\sqrt{2}}(|a_{1g}\rangle \mp i|e'_{g1}\rangle)$

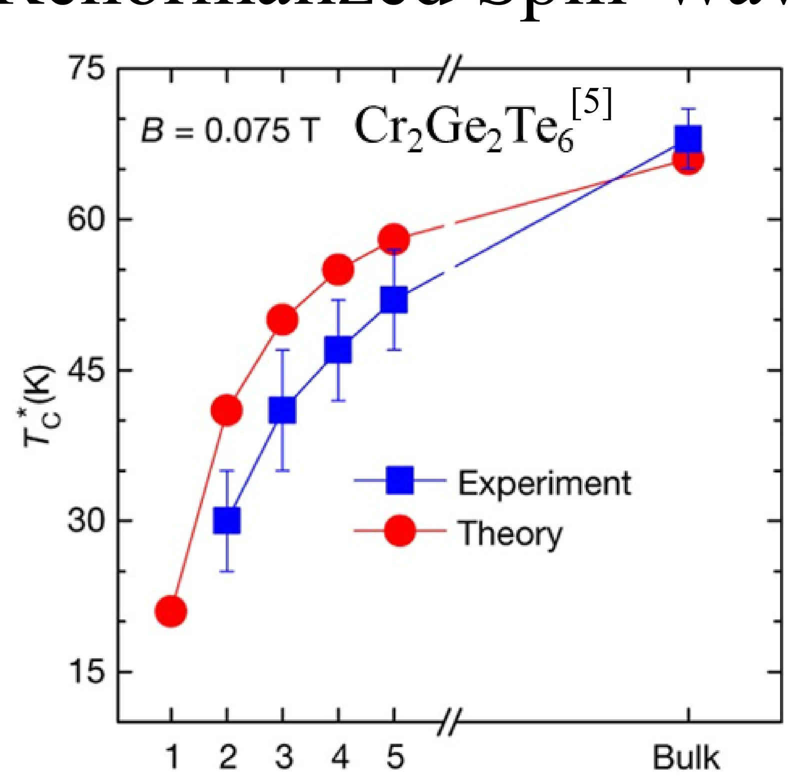
κ_1: L_z & L_x

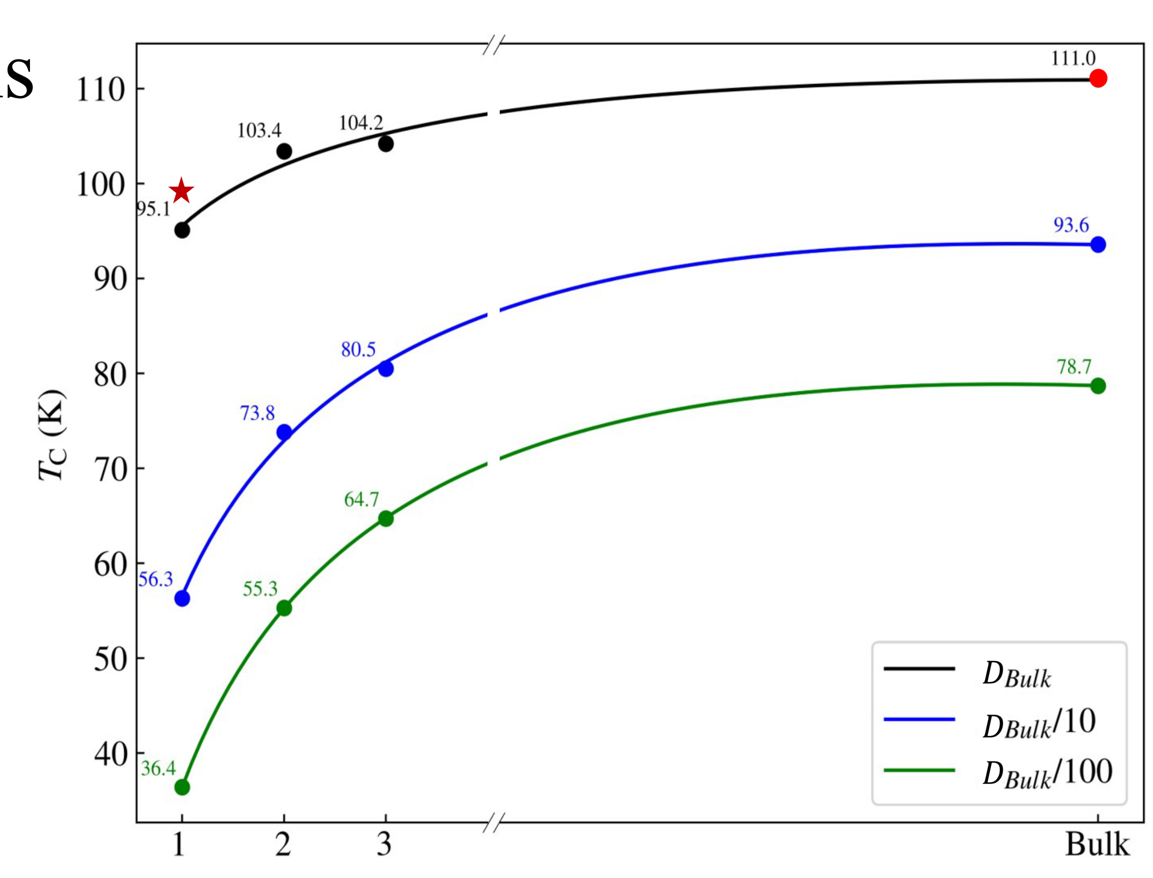
κ_2: L_z & L_y

κ_3: L_x & L_y



Renormalized Spin-Wave Simulations





large single-ion anisotropy(MAE) stabilizes the robust **FM T_C**

Reduced D significantly reduces T_C

★ $T_C \sim 99$ K for monolayer with $D = 6.74$ meV

A tensile strain would elevate T_C

Conclusions:

$a_{1g}^{\uparrow} L_{z-}^{\uparrow}$ Ising FM ground state with strong perpendicular magnetization for VI_3 of $R\bar{3}$ phase(with C_3 symmetry).

$\vec{M}$: $\theta \approx 30°(20°)$ for bulk(monolayer) VI_3 of $P\bar{1}$ phase (breaking C_3 symmetry). Exp[2]: $\theta \approx 36°$ for bulk
→ canted magnetization with reduced structural symmetry

strong perpendicular magnetic anisotropy effectively stabilizes the robust T_C

References:

[1] Kong et al., Adv. Mater. 31(17), 1808074 (2019)
[2] Hao et al., Chin. Phys. Lett. 38(9), 096101 (2021)
[3] Koriki et al., Phys. Rev. B 103(17), 174401 (2021)
[4] Lin et al., Nano Letters 21(21), 9180 (2021)
[5] Gong et al., Nature 546(7657), 265-269 (2017)