Finite temperature magnetic interactions from first principles

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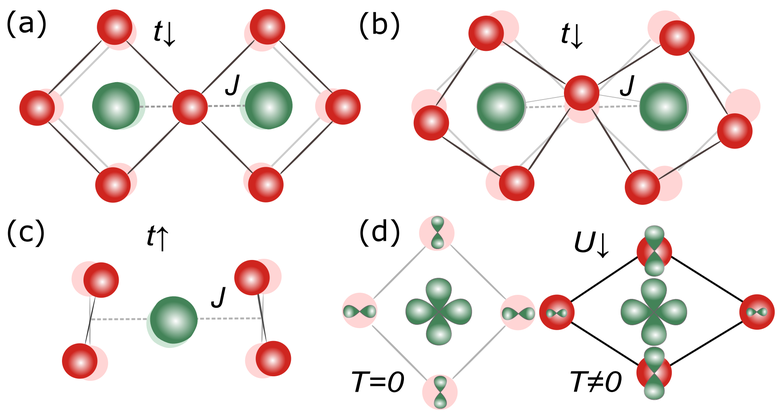
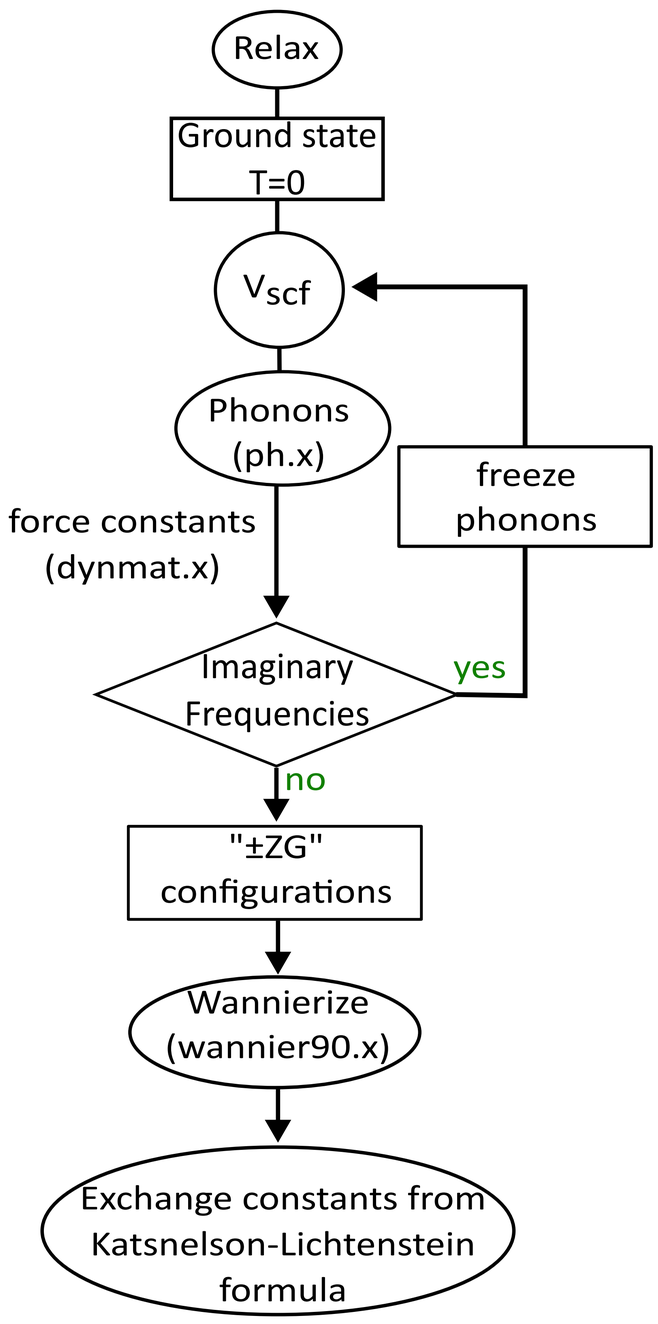
Motivation

Density functional theory has demonstrated remarkable predictive power in calculating magnetic properties at zero temperature. At finite temperatures, thermally excited phonons and structural phase transitions may affect magnetism. Efficient ab-initio methods to calculate the temperature dependence of magnetic exchange interactions are still lacking despite the importance of room temperature magnetism for applications. Here we introduce a special displacement method for calculating temperature-dependent magnetic interaction parameters from first principles and benchmark it on NiO. The approach allows to estimate magnetic interactions at finite temperatures with contributions from phonon-assisted quantum processes via two supercell calculations.



Workflow



References

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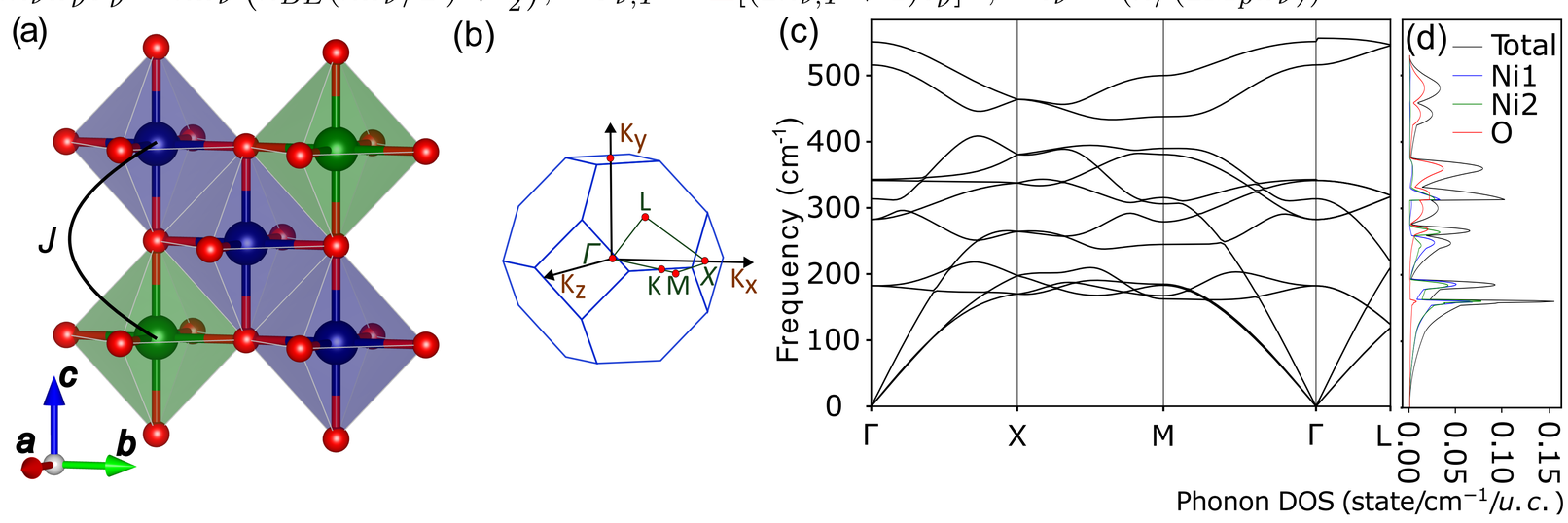
Sampling thermal phonons – Zacharias-Giustino method

Thermally excited phonons

$m_\nu\omega_\nu^2\sigma_\nu^2 = \hbar\omega_\nu\left(n_{BE}(\hbar\omega_\nu/T)+\frac{1}{2}\right), \quad \sigma_{\nu,T} = \pm[(2n_{\nu,T}+1)\ell_\nu^2]^{\frac{1}{2}}, \quad \ell_\nu = (\hbar/(2M_p\omega_\nu))^{1/2}$



Thermal lines

$\mathcal{T}_{\mathbf{S}}(T) = (S_1\sigma_{1(\nu,T)}(T), S_2\sigma_{2(\nu,T)}(T), \ldots, S_{3(N-1)}\sigma_{3(N-1)(\nu,T)}(T))$

T-dependent Heisenberg exchange interactions

Spin Hamiltonian

$\mathcal{H} = \frac{1}{2}\sum_{ij} J_{ij}(T)\mathbf{S}_i\cdot\mathbf{S}_j$

Temperature dependence of J

$J(T) = J_0 + \frac{1}{2}\sum_\nu \frac{\partial^2 J}{\partial x_\nu^2}\langle\sigma_{\nu,T}^2\rangle$

Katsnelson-Lichtenstein Method

$H = \sum_i H_{ii} + \sum_{i\neq j} H_{ij}, \quad H_{ii} \to e^{-\overrightarrow{v}_i\cdot\hat{J}_i} H_{ii} e^{-\overrightarrow{v}_i\cdot\hat{J}_i} = H_{ii} + i[H_{ii}, v_i\cdot\hat{J}_i]$

Second-order perturbation theory

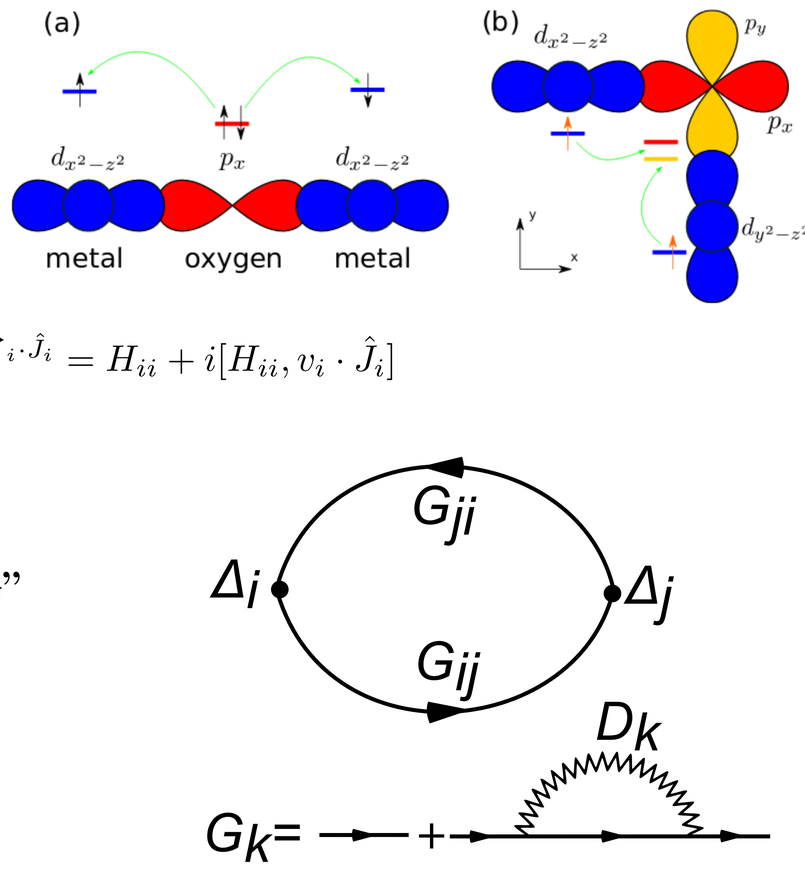
$\Delta E_{ij}^{(2)} = \frac{1}{2\pi}\mathrm{Im}\mathrm{Tr}(\Delta_i G_{ij}\Delta_j G_{ji})$

T-dependent J: average over "ZG" & "-ZG"

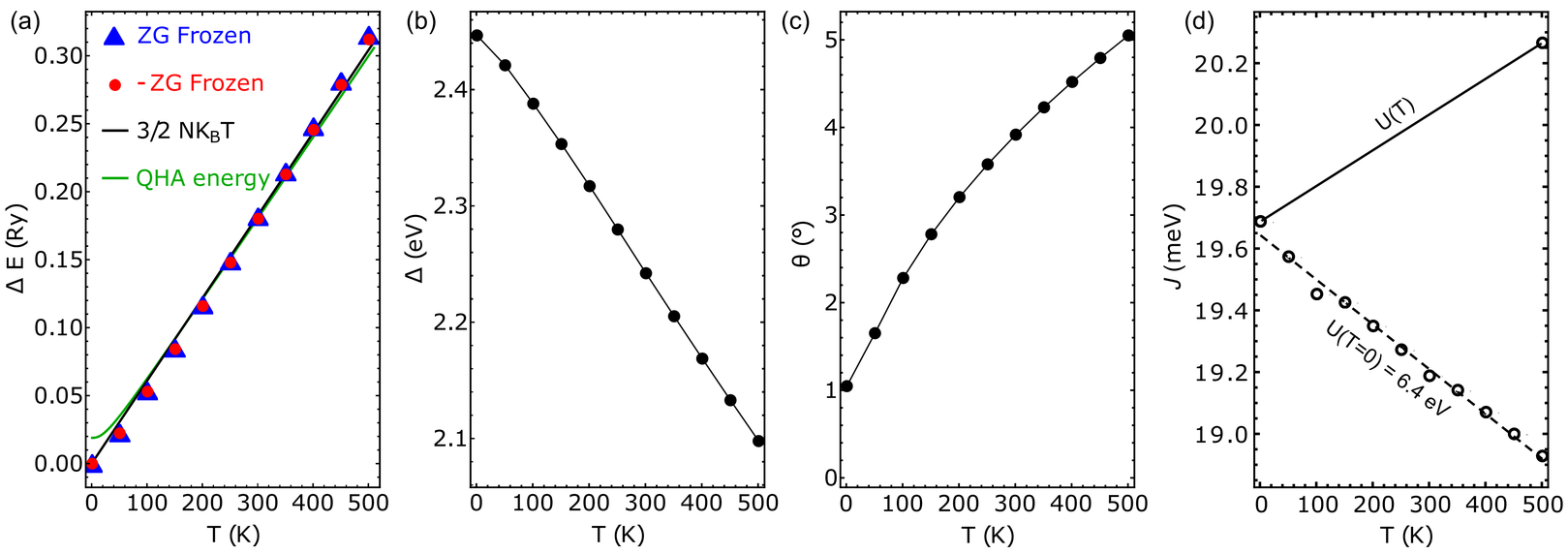
$J(T) = \frac{1}{2}\left(\langle J[\mathcal{T}_{S_i}]\rangle + \langle J[\mathcal{T}_{-S_i}]\rangle\right)$

Accounting for U corrections

$\frac{\Delta J}{J_0} = \left(\frac{(t'_\nu)^2}{t_0^2} + \frac{t''_{\nu\nu}}{t_0} - \frac{U''_{\nu\nu}}{2U_0} - 2\frac{t'_\nu}{t_0}\frac{U'_\nu}{U_0} + \frac{(U'_\nu)^2}{U_0^2}\right)\langle\sigma_\nu^2\rangle$



Results



Heisenberg exchange J(T) decreases in approximation of constant U. Self consistent treatment of U(T) could enhance J.

Conclusions and Outlook

- Modification of magnetic interactions by thermal phonons in solids
- Bond bending phonons reduce magnetic exchange constants with temperature
- Computationally affordable approach based on the special displacement method and Lichtenstein formula
  - ▷ Effects of thermal expansion
  - ▷ Inclusion of both thermal phonons and structural phase transitions
  - ▷ Soft halide-perovskites, 2D materials
  - ▷ Full self-consistency between phonons, displacements and U