Report: Effect of the electric field on the exchange interactions and magnon dispersion of NiI_2
by Andrey Rybakov, Dorye L. Esteras and José J. Baldoví

Electric field strength will be referenced as 50% for $E = 0.5$mV/Å and 100% for $E = 1$mV/Å. Corresponding structures will be referenced as e05 for $E = 0.5$mV/Å and e1 for $E = 1$mV/Å.

First of all there is the comparison of the structural displacement caused by the electric field. $\Delta r = |\vec{r}_0 - \vec{r}|$ (in Å) is defined with respect to the undistorted structure. Positive sign means displacement in the direction of the $\vec{b}$ lattice vector, negative – in the opposite direction.

Table 1

	undistorted	distorted	e05	e1
Ni, Δr, Å	0.0000	0.0705	0.0755	0.0776
I1, Δr, Å	0.0000	-0.0163	-0.0175	-0.0179
I2, Δr, Å	0.0000	-0.0163	-0.0175	-0.0179
$\Delta r(Ni)/\Delta r(I_1)$	-	4.3238	4.3238	4.3238

1 Magnon dispersion (method review)

Calculations are based on the [?, ?].

Spin Hamiltonian of the equation (1) is a starting point for the magnon dispersion calculation.

$$\hat{H} = -\sum_i \sum_\delta \hat{S}_i^T J_\delta \hat{S}_{i+\delta} \tag{1}$$

where J_δ is a 3×3 exchange matrix and $\hat{S}_i = (\hat{S}_i^x, \hat{S}_i^y, \hat{S}_i^z)^T$.

For treating of the helical state two rotations with rotation matrixes R and R_i are defined. R_i corresponds to the rotation of the «rotation frame» for each unit cell in the lattice and depends only on the lattice index i (or $i + \delta$). R rotates spin of the Ni atom within each rotation frame to the z direction of the rotation frame.Therefore, hellical state will be formed from the $\hat{S}_i''$ ferromagnetic state by applying those two rotation:

$$\hat{S}_i = R_i R \hat{S}_i''$$

Based on the matrix R two vectors are defined:

$$n_\alpha = R_{\alpha 3}$$

$$v_\alpha = R_{\alpha 1} + i R_{\alpha 2}$$

where first index corresponds to rows and second to columns and $\alpha = x, y, z$ $(1, 2, 3)$.

Standard linearized Holstein–Primakoff transformation is defined for the ferromagnetic configuration S_i'':

$$\hat{S}_i^{x''} = \sqrt{\frac{S}{2}}(\hat{b}_i + \hat{b}_i^\dagger)$$

$$\hat{S}_i^{y''} = \sqrt{\frac{S}{2}}(\hat{b}_i - \hat{b}_i^\dagger)$$

$$\hat{S}_i^{z\prime\prime} = S - \hat{b}_i^\dagger \hat{b}_i$$

then rotation R is applied which keeps the ferromagnetic configuration, but rotates the z axis to the new direction:

$$R\hat{S}'' = \sqrt{\frac{S}{2}}\overline{v}\hat{b}_i + \sqrt{\frac{S}{2}}v\hat{b}_i^\dagger + n(S - \hat{b}_i^\dagger \hat{b}_i)$$

where $\overline{v}$ means complex conjugate.

Substituting in the Hamiltonian and keeping only the meaningfull terms we get:

$$\hat{H} = -\sum_i \sum_\delta \left(\sqrt{\frac{S}{2}}\overline{v}\hat{b}_i + \sqrt{\frac{S}{2}}v\hat{b}_i^\dagger + n(S - \hat{b}_i^\dagger \hat{b}_i) \right)^T \cdot \\ \cdot R_i^T J_\delta R_{i+\delta} \cdot \\ \cdot \left(\sqrt{\frac{S}{2}}\overline{v}\hat{b}_{i+\delta} + \sqrt{\frac{S}{2}}v\hat{b}_{i+\delta}^\dagger + n(S - \hat{b}_{i+\delta}^\dagger \hat{b}_{i+\delta}) \right)$$

Infinite periodic spin lattice will not change under rotation R_i, since R_i rotates all spins in the unit cell equally:

$$\hat{H} = -\sum_i \sum_\delta \hat{S}_i^T J_\delta \hat{S}_{i+\delta} = -\sum_i \sum_\delta \hat{S}_i^T R_i^T J_\delta R_i \hat{S}_{i+\delta} => J_\delta = R_i^T J_\delta R_i$$

by redefining $J_\delta = R_i^T J_\delta R_{i+\delta}$ we get:

$$\hat{H} = -\sum_i \sum_\delta S^2 n^T J_\delta n - S n^T J_\delta n \hat{b}_i^\dagger \hat{b}_i - S n^T J_\delta n \hat{b}_{i+\delta}^\dagger \hat{b}_{i+\delta} + \\ + \frac{S}{2} v^\dagger J_\delta \overline{v} \hat{b}_i \hat{b}_{i+\delta} + \frac{S}{2} v^T J_\delta v \hat{b}_i^\dagger \hat{b}_{i+\delta}^\dagger + \\ + \frac{S}{2} v^\dagger J_\delta v \hat{b}_i \hat{b}_{i+\delta}^\dagger + \frac{S}{2} v^T J_\delta \overline{v} \hat{b}_i^\dagger \hat{b}_{i+\delta} + \dots$$

where $v^\dagger = \overline{v^T}$ is a hermitian conjugate. Further we omit the constant energy term $E_0 = S(S+1)n^T J_\delta n$.

This equation can be rewritten in a matrix form by defining $\hat{X}_i = (\hat{b}_i, \hat{b}_i^\dagger)^T$ ($\hat{X}_i^\dagger = (\hat{b}_i^\dagger, \hat{b}_i)$) and using commutation relation $[\hat{b}_i, \hat{b}_i^\dagger] = 1$:

$$\hat{H} = -\sum_i \sum_\delta \left(S(S+1) n^T J_\delta n - \hat{X}_i^\dagger h_1 \hat{X}_i + \hat{X}_i^\dagger h_2 \hat{X}_{i+\delta} \right)$$

where

$$h_1 = \begin{pmatrix} S n^T J_\delta n & 0 \\ 0 & S n^T J_\delta n \end{pmatrix}$$

$$h_2 = \begin{pmatrix} \frac{S}{2} v^T J_\delta \overline{v} & \frac{S}{2} v^T J_\delta v \\ \frac{S}{2} v^\dagger J_\delta \overline{v} & \frac{S}{2} v^\dagger J_\delta v \end{pmatrix}$$

Finally by applying the Fourier transformation

$$\hat{X}_i = \frac{1}{\sqrt{N}} \sum_k e^{ikr_i} \hat{X}_k$$

$$\hat{X}_i^\dagger = \frac{1}{\sqrt{N}} \sum_k e^{-ikr_i} \hat{X}_k^\dagger$$

where $\hat{X}_x = (\hat{b}_k, \hat{b}_{-k}^\dagger)^T$ ($\hat{X}_k^\dagger = (\hat{b}_k^\dagger, \hat{b}_{-k})$) and defining $J(k) = \sum_\delta J_\delta e^{-ik\delta}$ we get:

$$\hat{H} = -\sum_k \hat{X}_k^\dagger h(k) \hat{X}_k$$

where

$$h(k) = \begin{pmatrix} \frac{S}{2} v^T J(-k) \overline{v} - S n^T J(0) n & \frac{S}{2} v^T J(-k) v \\ \frac{S}{2} v^\dagger J(-k) \overline{v} & \frac{S}{2} v^\dagger J(-k) v - S n^T J(0) n \end{pmatrix}$$

Since $J_\delta^T = J_{-\delta}$ the following holds:

$$\overline{J(k)^T} = \sum_\delta J_\delta^T e^{ik\delta} = \sum_{-\delta} J_{-\delta} e^{-ik(-\delta)} = \sum_\delta J_\delta e^{-ik\delta} = J(k)$$

therefore the matrix $h(k)$ is hermitian and has the form:

$$h(k) = \begin{pmatrix} A(k) - C & B(k) \\ B(k)^\dagger & \overline{A(-k)} - C \end{pmatrix}$$

where

$$A(k) = \frac{S}{2} v^T J(-k) \overline{v}$$

$$B(k) = \frac{S}{2} v^T J(-k) v$$

$$C = S n^T J(0) n$$

The last step is to find new collective creation and annihilation operators $\hat{Y} = (\hat{c}_k, \hat{c}_{-k}^\dagger)^T$ which obeys the bosonic commutation relations and diagonalizes the Hamiltonian:

$$\hat{H} = -\sum_k \hat{Y}_k^\dagger \Omega(k) \hat{Y}_k$$

where

$$\Omega(k) = \begin{pmatrix} \omega_1(k) & 0 \\ 0 & \omega_2(k) \end{pmatrix}$$

We are looking for the canonical transformation S: $\hat{X} = S\hat{Y}$.

$$\hat{H} = -\sum_k \hat{X}_k^\dagger h(k) \hat{X}_k = -\sum_k \hat{Y}_k^\dagger S^\dagger h(k) S \hat{Y}_k = -\sum_k \hat{Y}_k^\dagger \Omega(k) \hat{Y}_k$$

therefore

$$\Omega(k) = S^\dagger h(k) S \tag{2}$$

Here we recall commutation relations in a matrix form for the bosonic operators:

$$[\hat{X}_k, \hat{X}_k^\dagger] = \hat{X}_k \hat{X}_k^\dagger - (\hat{X}_k^{\dagger T} \hat{X}_k^T)^T = g = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

same holds for $\hat{Y}_k$.

$$g = [\hat{X}_k, \hat{X}_k^\dagger] = [S\hat{Y}_k, \hat{Y}_k^\dagger S^\dagger] = S\hat{Y}_k\hat{Y}_k^\dagger S^\dagger - ((S\hat{Y}_k)^{\dagger T}(\hat{Y}_k S)^T)^T =$$
$$= S\hat{Y}_k\hat{Y}_k^\dagger S^\dagger - S(\hat{Y}_k^{\dagger T}\hat{Y}_k^T)^T S^\dagger = S\left(\hat{Y}_k\hat{Y}_k^\dagger - (\hat{Y}_k^{\dagger T}\hat{Y}_k^T)^T\right) S^\dagger = SgS^\dagger$$

therefore

$$S^\dagger = g^{-1} S^{-1} g$$

By substituting this relation in the (2) we get:

$$h(k)S = g^{-1} S g \Omega(k) = gSg\Omega(k)$$

which could be solved in order to obtain solutions for ω_1 and ω_2

$$\omega_1(k) = \frac{A(k) - \overline{A(-k)} \pm \sqrt{(A(k) + \overline{A(-k)} - 2C)^2 - 4B(k)^\dagger B(k)}}{2}$$
$$\omega_2(k) = \frac{\overline{A(-k)} - A(k) \pm \sqrt{(A(k) + \overline{A(-k)} - 2C)^2 - 4B(k)^\dagger B(k)}}{2}$$

Lets define $D = (A(k) + \overline{A(-k)} - 2C)^2 - 4B^\dagger B$ and $\delta_k = A(k) - \overline{A(-k)}$ and rewrite those equation for ω in the following way:

$$\omega_1(k) = \frac{\pm\sqrt{D} + \delta_k}{2}$$
$$\omega_2(k) = \frac{\pm\sqrt{D} - \delta_k}{2}$$

From this form we can see that the real frequencies which will correspond to the wave with k and $-k$ wave-vectors are the ones with the positive sign before $\sqrt{D}$ (take for example the simple 1-D chain of spins with isotropic exchange: $\omega_1 = \omega_2 = 2J_{iso}(1 - cos(ka)))$ and δ_k is the difference between magnon modes with opposite k-vectors.

$A(k)$, C and $B(k)^\dagger B(k)$ are real, thus δ_k and D are real, but ω will have imaginary part if $D < 0$.

2 Occupation «smearing»

2.1 Band structure and wannier fit

The quality of the wannier fit is analysed through the average (η) and maximum (η_{max}) «band distance», which are defined as:

$$\eta = \max_{nk}(|\varepsilon_{nk}^{DFT} - \varepsilon_{nk}^{Wannier}|)$$

$$\eta_{max} = \sqrt{\frac{1}{N}\sum_{nk}(\varepsilon_{nk}^{DFT} - \varepsilon_{nk}^{Wannier})^2}$$

In Fig. ?? the dependencies of the band distance are present for the distorted structure (for the rest 3 structure the values are very similar). For each structure there are three calculations: with magnetisation along x, y and z axes. z axis is out of plane. The values indicate good quality of the Wannier fit.

Band gap obtained in the calculations for different U may be used as a criteria for choosing particular U value. In fig. ?? the evolution of the band gap is presented for all four structures.

2.2 Exchange

Isotropic exchange interactions for all 4 structures and Hubbard $U = 0 - 5$ eV are provided in tables 2, 3, 4.

Table 2: Isotropic exchange interaction $J_1(4)$ and $J_1(2)$ vs Hubbard U.

$J_1(4)$, meV					$J_1(2)$, meV				
U, eV	undist	dist	e05	e1	U, eV	undist	dist	e05	e1
0	3.82	3.82	3.82	3.82	0	3.82	3.88	3.90	3.90
1	3.54	3.53	3.53	3.53	1	3.54	3.60	3.61	3.61
2	3.32	3.31	3.29	3.29	2	3.32	3.38	3.38	3.38
3	3.15	3.11	3.11	3.11	3	3.15	3.17	3.19	3.19
4	2.99	2.95	2.94	2.94	4	2.99	3.02	3.01	3.02
5	2.81	2.81	2.81	2.79	5	2.81	2.87	2.88	2.86

Table 3: Isotropic exchange interaction $J_2(4)$ and $J_2(2)$ vs Hubbard U.

$J_2(4)$, meV					$J_2(2)$, meV				
U, eV	undist	dist	e05	e1	U, eV	undist	dist	e05	e1
0	-0.02	-0.02	-0.02	-0.02	0	-0.02	-0.03	-0.04	-0.04
1	0.02	0.02	0.02	0.02	1	0.02	0.01	0.01	0.01
2	0.04	0.03	0.03	0.03	2	0.04	0.03	0.02	0.02
3	0.04	0.04	0.04	0.04	3	0.04	0.03	0.03	0.03
4	0.05	0.04	0.04	0.04	4	0.05	0.04	0.04	0.04
5	0.05	0.05	0.05	0.05	5	0.05	0.04	0.04	0.04

Table 4: Isotropic exchange interaction $J_3(4)$ and $J_3(2)$ vs Hubbard U.

$J_3(4)$, meV				
U, eV	undist	dist	e05	e1
0	-4.29	-4.22	-4.21	-4.20
1	-3.86	-3.81	-3.80	-3.80
2	-3.39	-3.39	-3.38	-3.37
3	-3.06	-3.03	-3.02	-3.02
4	-2.76	-2.75	-2.74	-2.74
5	-2.48	-2.47	-2.46	-2.46

$J_3(2)$, meV				
U, eV	undist	dist	e05	e1
0	-4.29	-4.14	-4.11	-4.10
1	-3.86	-3.74	-3.72	-3.71
2	-3.39	-3.33	-3.30	-3.30
3	-3.06	-2.97	-2.96	-2.95
4	-2.76	-2.70	-2.68	-2.68
5	-2.48	-2.43	-2.42	-2.42

3 Occupation «fixed»

We tried to include electric field in the DFT calculations as explained in [**?**, **?**], which requires the calculations with fixed occupations in QE. However the Wannier fit in that case does not reproduce the electronic band structure (see fig. **??**), thus we can not rely on the computed exchange values in that case. Currently we are trying to include the effect of an electric field in the calculation.

References

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