Enhanced magnetic anisotropy and Curie temperature of

NiI$_2$ monolayer by applying strain: first-principles study

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S1. The details of self-consistent calculation for U.

According to the linear response approach proposed by Cococcioni and Gironcoli,[1] U is determined by the difference between the screened and bare second derivative of the energy with respect to localized state occupations λ^I at site *I*. This can be given as:

$$U = \frac{\partial^2 E[\{\lambda^I\}]}{\partial(\lambda^I)^2} - \frac{\partial^2 E_0[\{\lambda^I\}]}{\partial(\lambda^I)^2}$$

Applying localized potential shifts to the d levels of the Ni atoms to excite charge fluctuation on their orbitals, and solving the Kohn–Sham equations self-consistently, an occupation-dependent energy functional can be obtained:

$$E[\{\lambda^I\}] = \min_{\beta_I}\left\{E[\{\beta_I\}] - \sum_I \beta_I \lambda^I\right\}$$

Thus

$$\frac{\partial E[\{\lambda^I\}]}{\partial \lambda^I} = -\beta_I(\{\lambda^I\}), \quad \frac{\partial^2 E[\{\lambda^I\}]}{\partial(\lambda^I)^2} = -\frac{\partial \beta_I(\{\lambda^I\})}{\partial \lambda^I},$$

Using β_I as the perturbation parameter, the effective interaction parameter U of site I can then be written as,

$$U = \frac{\partial \beta_{I,0}}{\partial \lambda_I} - \frac{\partial \beta_I}{\partial \lambda_I} = [\chi_0^{-1} - \chi^{-1}]^I$$

In the above derivation, U is calculated from the GGA ground state; it should be consistently obtained from the GGA+U ground state itself, which may be especially relevant when GGA and GGA+U differ qualitatively. Thus, Scherlis   et al. have

identified that the electronic terms in the GGA+U functional have quadratic dependence on the occupations: [2]

$$E_{quad} = \frac{U_{scf}}{2} \sum_I [\sum_i \lambda_i^I (\sum_j \lambda_j^I - 1)] + \frac{U_{in}}{2} \sum_I \sum_i \lambda_i^I (1 - \lambda_i^I)$$

Where, the first term is the contribution already contained in the standard GGA functional; the second term is the customary ''+U'' correction. Thus, U_{scf} represents the effective on-site electron-electron interaction already present in the GGA energy functional for the GGA+U ground state when U is chosen to be U_{in}. The second derivative of E_{quad} with respect to λ_i^I also corresponds to the U_{out} obtained from linear-response:

$$U_{out} = \frac{d^2 E_{quad}}{d(\lambda_T^I)^2} = U_{scf} - \frac{U_{in}}{m}$$

Where, m can be interpreted as an effective degeneracy of the orbitals whose population is changing during the perturbation. U_{out} is linear in U_{in} for the relevant range of U_{in}–U_{scf}. From a few linear-response calculations for different U_{in} ground states, we can extract the U_{scf} that should be used for the NiI$_2$ monolayer. The self-consistent determination of U is performed by using the quantum espresso (QE) code[3] with the GGA (PBE) exchange–correlation functional and PAW pseudopotentials. The wave-function and electronic density cut-off energies are 60 Ry and 400 Ry, respectively. Fig. S1 plots U_{out} as a function of U_{in}, which shows a good linear relationship. It can be seen that the extrapolated U for NiI$_2$ monolayer is 7.33 eV.
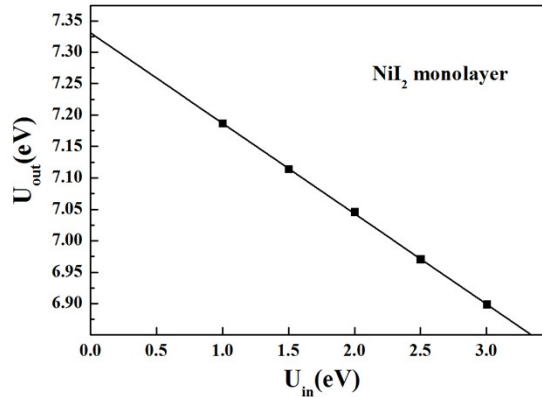

Fig. S1 Linear response U$_{out}$ calculated from the U$_{in}$ ground state of NiI$_2$ monolayer.

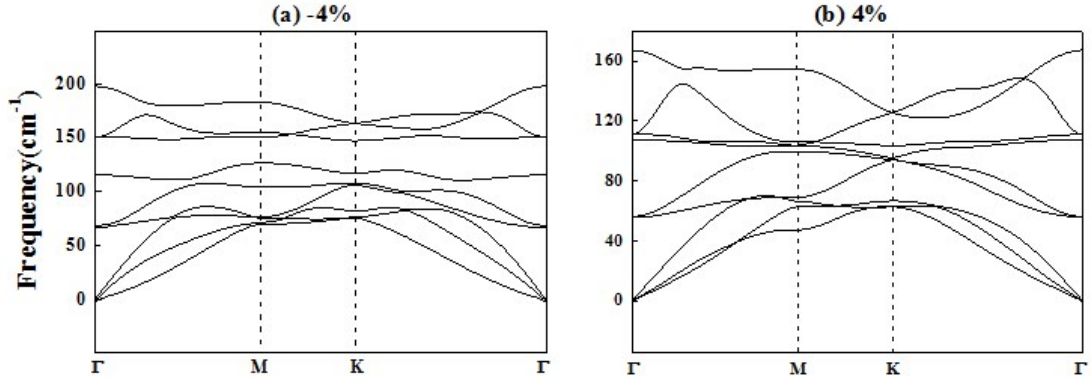**S2. The phonon spectra of the NiI$_2$ monolayer under the -4% and 4% strains.**

Fig. S2 The phonon spectra of the NiI$_2$ monolayer under the -4% and 4% strains.

S3. MD movies of NiI$_2$ monolayer under the 0%, -4% and 4% strains.

·0%.wmv represents the NiI$_2$ monolayer under 0% strian at T=300K.

·-4%.wmv represents the NiI$_2$ monolayer under -4% strian at T=300K.

·4%.wmv represents the NiI$_2$ monolayer under 4% strian at T=300K.

S3. The details of calculating exchange parameter J from the energy difference between FM and AFM states(ΔE).

The Hamiltonian can be expressed as：

$$H = -\sum_{i,j} JS_i S_j$$

Where J is the nearest-neighboring exchange parameter. From Fig. 2(b) and 2(c) of the main text, we can estimate J from the following equation for the NiI$_2$ monolayer. [4]

We construct a 2×2 supercell:

$$E(FM) = -(12J)S^2$$

$$E(AFM) = -(-4J)S^2$$

$$\Delta E = E(AFM) - E(FM) = 16JS^2$$

$$J = \frac{\Delta E}{16S^2}$$

References

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