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Magnetic Exchange Interactions and Estimation of T_{N} in $CsNiF_3$ from First Principles

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The $CsNiF_3$ has been for long time studied as a prototype of quasi-1D planar ferromagnetic system. At very low temperature the studied system is insulator and therefore the magnetic exchange interactions should decay very rapidly. We treated the magnetic exchange coupling within the Heisenberg model for the nearest neighbor interaction between the antiferromagnetically coupled Ni-chains. The influence of up to the second-nearest neighbors on the ferromagnetic exchange coupling along the Ni-chains was determined. The exchange interactions were calculated for the experimental volume by the density functional theory, within the all-electron approach using the local density approximation for the exchange and correlation. The Néel temperature was calculated by means of the mean-field theory and by the random-phase approximation method.

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1. Introduction

The $CsNiF_3$ serves as thoroughly studied prototype of 1D linear-chain ferromagnet that only develops some 3D ordering under the $T_{\mathrm{N}} = 2.7$ K, that was first measured in [1]. The hexagonal structure with lattice parameters $a = 6.236$ Å, $c = 5.225$ Å [1] (space group 194, $P6_3/mmc$, Pearson symbol hP6) consists of 10 atoms per unit cell with the Wyckoff position 2d (Cs), 2a (Ni) and 6h (F). The Ni-chains are formed under 80 K along the crystal c-axis (z-axis), while within the $a{-}b$ ($x{-}y$) plane they are separated by the large Cs ions. Within this plane the onset of antiferromagnetism appears. Many experimental studies have investigated

different magnetic properties, for review see [3, 4], ultrasonic study of magnetic phases [5], spin waves [6], magnon dispersions [7], excitation spectrum in the magnetic field within [8] and perpendicular [9] to the easy plane and etc. Such 3D insulating systems occurring at very low temperatures could be very well studied using state-of-art *ab initio* methods. As the starting point we investigated the strength of the exchange interaction and compared them to the recently revised experimental studies [7, 10]. Moreover, the critical temperature (Néel temperature) was calculated using the mean-field theory and random phase approximation.

2. Methodology

The electronic structure was calculated using the full-potential linearized plane wave code WIEN2k [11] employing the local density approximation (LDA) for the exchange and correlation term. The muffin-tin radii were set to 2.5 (Cs), 2.0 (Ni) and 1.6 (F) atomic units, the cut-off energy expressed as the product of the muffin-tin radii and the maximum plane wave vector was equal to 7 and the 60 $\boldsymbol{k}$-points for the whole Brillouin zone in order to guarantee the total energy error bar better than 0.00001 Ry per unit cell.

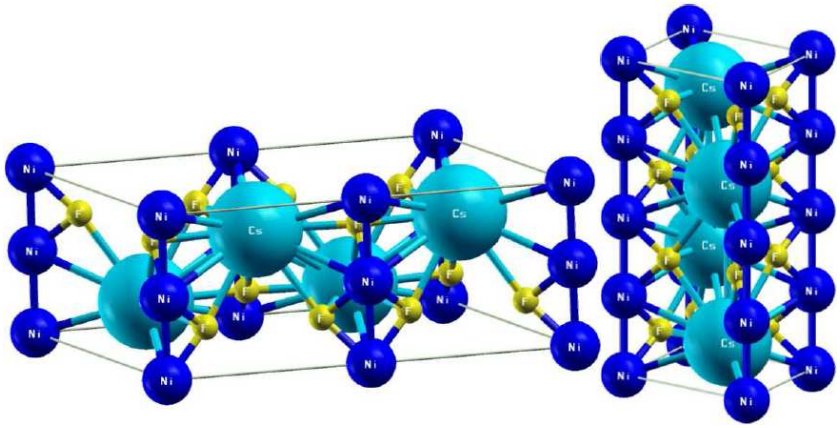

Fig. 1. The models of the $CsNiF_3$ compound to calculate spin-flip configurations for the first, $2 \times 1 \times 1$ supercell (left), and up to second, $1 \times 1 \times 2$ supercell (right), nearest--neighbor exchange interactions.

The intra and inter Ni-chains exchange interactions were calculated from the total energies for different magnetic configurations using an approach presented in Ref. [12]. For this task we employed $2 \times 1 \times 1$ and $1 \times 1 \times 2$ supercells, see Fig. 1. The obtained exchange interactions were used to construct the classical Heisenberg model, from which we evaluated the critical temperature. Two approximations were used — the mean field approximation (MFA) and multi-sublattice random phase approximation (RPA) [13].

3. Results and discussion

By evaluating the total energies of the single and double site flipping of the spins and comparing them to the configuration with all spins parallel (ferromagnetic solution) we obtained the exchange interaction strength $J = 1.83$ meV (21.3 K) between nearest neighbor Ni-atom (along c-axis), whereas the exchange interaction between nearest chains is very weak, $J' = -0.01$ meV (0.1 K). The negative resulting J indicates the antiferromagnetic, whereas the positive J, the ferromagnetic coupling. The ferromagnetic exchange interaction along the Ni-atom chain is in excellent agreement with the experimental value of 23.6 K [7, 10], but for the antiferromagnetic coupling between Ni-chains the calculated value is about twice the experimental one $J' = -0.05$ K [10]. However, such small exchange interactions are near to the edge of accuracy of total energy calculations. Nevertheless, the ratio of the intra- to the inter-exchange interaction constants is of the same order as in the experiment. From the second supercell considered ($1 \times 1 \times 2$) we found out that the second nearest neighbor atom contribution (along the Ni-chain, c-axis) to the ferromagnetic exchange interaction is very weak, $J(2\text{NN}) = 0.005$ meV (0.06 K). Thus, it is safe to consider only the first nearest neighbor interactions in $CsNiF_3$ compound. The critical (Néel) temperature was estimated by two different approaches, the MFA and RPA, using the calculated exchange interactions. The MFA leads to the value of $T_C = 38.8$ K, strongly overestimating the experimental critical temperature. The reason is that MFA neglects the $\boldsymbol{q}$-dependence of the exchange interactions (here the weak interchain coupling is averaged with stronger intrachain interactions). The Néel temperature calculated within the RPA $T_N = 6.9$ K is a few K above the measured one T_N [1]. The exact estimation of T_N within the Heisenberg model would be possible using the time and resources more demanding Monte Carlo approach and it would most likely increase the RPA estimate. The remaining disagreement with respect to the experiment can be probably explained by the presence of crystal field effect, that is of the same order of magnitude compared to the strongest exchange interaction J [7]. Crystal field effects typically decrease estimated critical temperatures [14].

4. Conclusion

To the best of our knowledge the very first full-potential all-electron *ab initio* calculation of $CsNiF_3$ was performed employing the LDA for the exchange-correlation term. The magnetic exchange interactions were evaluated for the two different models to use the Heisenberg Hamiltonian. The $2 \times 1 \times 1$ supercell calculations revealed the correct ferromagnetic exchange interaction along the Ni-atoms and the very weak but finite antiferromagnetic ordering among the Ni-chains, supporting the 3D ordering under T_N. The $1 \times 1 \times 2$ supercell calculations have confirmed that the second-nearest atom contribution to the exchange interaction along the Ni-atoms chain is very weak. Moreover, the critical Néel temperature

was calculated using the strength of calculated exchange interaction according to the mean-field and random-phase approximation.

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