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Report:

The relationship between hyperfine interactions and magnetism in fully ordered double-perovskites A_2NiMnO_6 (A = Sc, In, Tl) is investigated by nuclear resonance scattering (NRS) with the ^{61}Ni transition, and compared to neutron diffraction measurements [1,2]. In these perovskite-like oxides, the increased octahedral ($Ni^{2+}O_6$) and ($Mn^{4+}O_6$) tilting associated with decreasing A-site ionic radius ($Tl^{3+} \rightarrow In^{3+} \rightarrow Sc^{3}$) is a key ingredient in changing the sign of the nearest-neighbor (NN) Ni-O-Mn magnetic interactions.

In the case of Sc_2NiMnO_6 with antiferromagnetic NN-interactions, exhibiting two magnetic transitions at 35 K and 17 K, the magnitude and sign of the magnetic hyperfine field (B_{hf}) on ^{61}Ni nuclei is determined using NFS measurements (Fig. 1). The measurements of Sc_2NiMnO_6 at 4.6, 14.9, 23.8 and 33.2 K were done at the magnetic region and above Neel point at 39.7 K (Fig. 1a-e). The 39.7 K spectrum (Fig. 1e) fit suggests that there is no measurable quadrupole splitting in the sample. The effective thickness of the sample was obtained from dynamical beats as L=5.68. $L=\mu_n * z * f_{LM}$, where $\mu_n = 0.00868\ \mu m^{-1}$ – nuclear absorption factor, z – sample thickness, f_{LM} – Lamb-Mossbauer factor. Using z=2000 μm, obtain $f_{LM} = 0.33$ (Fig 2b).

The spectra below Neel temperature were fitted with 2 models: (1) one site with isotopically distributed magnetic field, (2) the site with magnetic splitting and the site with single line (non-magnetic). The second model was used to check possible transition of the Ni to the paramagnetic state above anomaly temperature around 17K. The parameters of the fit were scaling factor I_0, time zero dt, Lamb-Mossbauer factor f_{LM} (describing line broadening), hyperfine magnetic field B_{hf}, and the relative contribution of the non-magnetic site a2 (see figure 2c). All fits were done using Motif software [3]. The fit of the data by these two models are shown for several temperatures in the Fig.1a-d, where red color corresponds to the first model, blue color – to the second model. Obtain the both models fit quite well measured data. Presence of the non-magnetic site is completely absent below 19K and is of the order of 1-2% between 20 and 40 K. Temperature dependence of the hyperfine magnetic field, Lamb-Mossbauer factor and contribution of the non-magnetic site are shown in Fig.3

Figure 3 shows NFS spectrum of Sc_2NiMnO_6 sample was measured at 5 K with applied vertical field of B_{ex} = 6 T. This spectrum was fitted using 2 models: (1) assuming that all magnetic moments aligned along the field (ferromagnetic alignment). In this case, one would expect that only lines with Δm=0 will be present in the spectrum. Parameters of the fit here are total hyperfine magnetic field and additional broadening; (2) – assuming that field does not affect isotropic distribution of magnetic moments, model with integrates over isotropic distribution with additional external field of 6T in vertical direction (antiferromagnetic alignment).

The small absolute value of the field $B_{hf,2.5K} \approx 6.0$ T on Ni^{2+} ions in the octahedral oxygen coordination is explained by the large positive orbital contribution ($B_L > 0$) due to the 3*d*-4*p* orbital mixing via spin-orbit coupling. The negative sign of B_{hf} in the external field underlines that the core polarization ($B_F \approx -13$ T) is the most important partial contribution to the experimental magnetic hyperfine field. The temperature evolution of the reduced hyperfine fields $B_{hf}(T)$ is reproduced by the Brillouin function (Fig. 2a) with $S = 1$ and magnetic transition temperature of 38(2) K, that is incompatible with the earlier assumption [1] that the low temperature transition at $T_{N2} \approx 17$ K arises from the antiferromagnetic ordering in Ni^{2+} sublattice. The significantly lower values of the hyperfine field in the ferromagnet Tl_2NiMnO_6 ($B_{hf,5K} \approx$ 1.7 T) and In_2NiMnO_6 ($B_{hf,5K} \approx 2.1$ T) (Fig. 2a) with a cycloidal magnetic structure [2] are entirely associated with the supertransferred hyperfine field (B_{STHF}) from the nearest Mn^{4+} neighbors via an intermediate O^{2-} ions. Taking into account the angular dependence of the $B_{STHF}(\vartheta)$ field on the Ni-O-Mn bond angle ϑ, we have shown that, in opposite to Sc_2NiMnO_6, the B_{STHF} field in A = In, Tl perovskites have the positive sign, thus drastically reducing the resulting B_{hf} value.

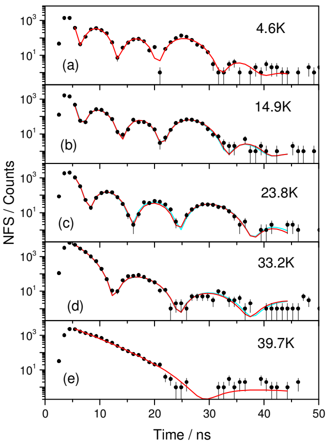

Figure 3. NFS spectra of Sc_2NiMnO_6 at several temperatures. Red and blue colors denote fit by models (1) and (2)

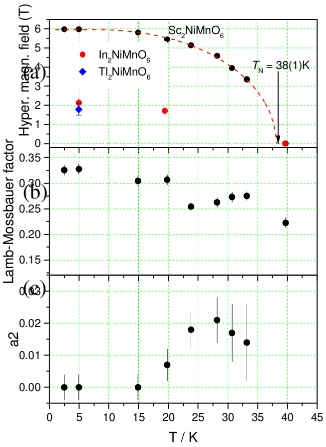

Figure 3. Temperature dependence of B_{hf}, f_{LM} and relative contribution of non-magnetic site for Sc_2NiMnO_6

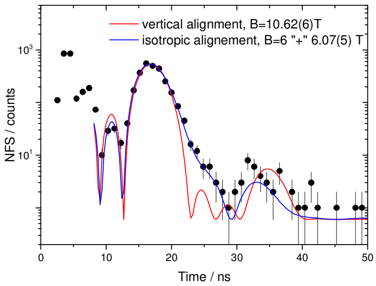

Figure 3. NFS spectrum of Sc_2NiMnO_6 in external 6T field. Red and blue colors are fit with 2 models presented in the text.

References

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