^{61}Ni Nuclear Resonance Scattering Study of Magnetic Hyperfine Interactons in the Double-Perovskites A_2NiMnO_6 (A = Sc, In, Tl)

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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 with and without external magnetic field B_{ex} = 5 T. The small absolute value of the field $B_{hf,2.5K} \approx 6.1$ 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 ($B_F \approx -40$ 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 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) 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.

The European Synchrotron Radiation Facility is acknowledged for provision of synchrotron radiation facilities at beamline ID18.

References

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