Effect of rare-earth ions on magnetic properties and spin excitations in manganites

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Rare-earth (R) ions can significantly affect magnetic properties of pure and doped manganites $RMnO_3$ with an orthorhombic perovskite structure (Pbnm) by means of a change of the R-ion ionic radii, influencing the Mn–O–Mn angle and thus the Mn–Mn exchange. This results in decreasing of the Neel temperature, T_N, from 140 K for $LaMnO_3$ to 40 K for $TbMnO_3$ and corresponding transformation of the magnetic structure from an antiferromagnetic (AF) layer A-type ordering with a weak ferromagnetism (A_yF_z) (La, Pr, Nd) [1] to an incommensurate (IC) sinusoidal structure (Tb) [2]. The other effect of the R ions is their direct (para)magnetic contribution at low temperatures which results in a significant anisotropy of magnetic properties as was recently observed in $PrMnO_3$ and $NdMnO_3$ [3].

In this work we studied magnetic properties and spin excitations in the orthorhombic $RMnO_3$ with smaller R-ion ionic radii (R = Sm, Eu, Gd). Single crystals of the corresponding manganites were grown by the floating zone method. X-ray powder diffraction measurements showed that the crystals were single-phase with an orthorhombic structure. The observed strong magnetic and dielectric anisotropy indicates that the crystals are practically untwined. Magnetization and susceptibility were studied in the magnetic fields up to 5 T at $T = 2-300$ K. Transmission spectra of $RMnO_3$ were measured in the frequency range $\nu = 2-33\,\mathrm{cm}^{-1}$ by means of quasi-optical submillimeter (SBMM) backward-wave-oscillator technique.

$SmMnO_3$. Asignificant anisotropy of the magnetic susceptibility ($\chi_a > \chi_c > \chi_b$) due to Sm contribution and a noticeable temperature dependence of the spontaneous magnetization along the *c*-axis with a compensation point at 9 K were observed in $SmMnO_3$ (Figs. 1a and b), which indicates an antiparallel orientation of the Mn weak ferromagnetic moment and Sm one induced by Sm–Mn exchange interaction. Four modes, $\nu_{F,AF}$ and $\nu_{R1,R2}$, were observed in the SBMM transmission spectra of the $SmMnO_3$ below $T_N \approx 60$ K (Fig. 2). Two of them were identified as AFMR modes of the Mn-subsystem while other two ($T < 35$ K)—as the R-modes determined by the magnetodipolar transitions inside the exchange-

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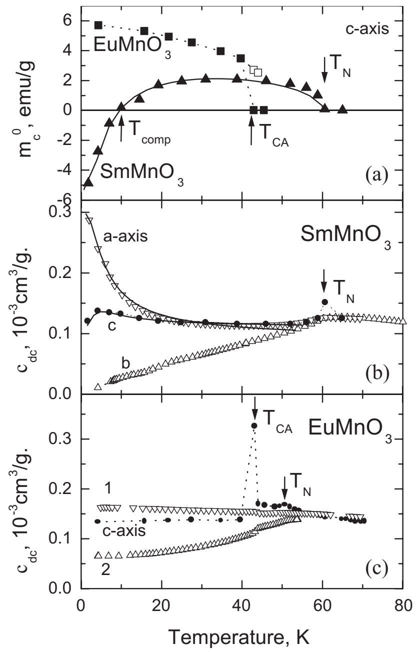

Fig. 1. Temperature dependence of the spontaneous magnetization for $SmMnO_3$ (triangles) and $EuMnO_3$ (squares) single crystals along *c*-axis (open squares—spontaneous magnetization in the field-induced canted phase) (a) and DC susceptibility for $SmMnO_3$ (b) and $EuMnO_3$ (c) along three crystallographic axes. Points—experiment, solid lines—theory, dotted lines—guides for the eye. Arrows indicate the Neel temperature, the temperature of the transition to the canted AF phase and the compensation point.

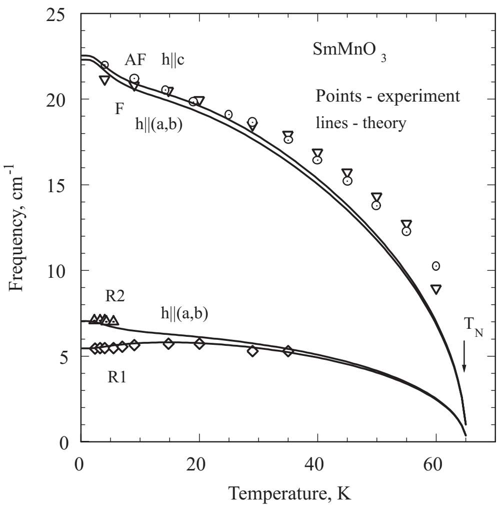

Fig. 2. Temperature dependence of the resonance frequencies of quasiferromagnetic (F, triangles) and quasiantiferromagnetic (AF, circles) AFMR Mn-modes and two Sm-modes (R1, R2) in $SmMnO_3$: Points—experiment, lines—theory.

split ground Sm^{3+} doublet. Self-consistent description of both static and dynamic $SmMnO_3$ properties were performed (Figs. 1 and 2) and parameters of magnetic interactions were determined.

$EuMnO_3$. The onset of an AF ordering was observed at $T_N \approx 50\,K$ as a weak peak in $\chi_c(T)$ followed by a strong peak at $T_{CA} \approx 43\,K$, accompanied by a sharp appearance of a weak ferromagnetic moment along the *c*-axis (Figs. 1a and c). No spontaneous magnetic moment was observed between T_N and T_{CA}, however, it is induced by the magnetic field ~ 0.5–$1\,T$ along the *c*-axis (open squares in Fig. 1a). Taking into account a tendency of appearing of a sinusoidal structure instead of a canted A-type structure with smaller R-ion ionic radii (Tb, Ho) [2,4], we suggest an IC AF state in $EuMnO_3$ between T_N and T_{CA}, which competes with the canted AF state and is suppressed below T_{CA}. In the SBMM spectra we observed an AFMR in Mn subsystem below 45 K with a behavior similar to that in $SmMnO_3$.

$GdMnO_3$. Here we observed further decrease of the transition temperature to the canted state ($T_{CA} \sim 20\,K$). On the other hand no visible anomalies in $\chi_c(T)$ were observed above T_{CA} at the expected transition to the AF incommensurate phase, perhaps, due to a large background of the Gd subsystem. However, a well-defined field induced transition accompanied by a jump of magnetization were observed above T_{CA}, similar to $EuMnO_3$. It indicates the existence of the AF incommensurate state above T_{CA} which is suppressed by the field. At $T \sim 7\,K$ a maximum of the susceptibility and a subsequent decrease of the spontaneous magnetization were observed which could be related to the AF ordering of the Gd subsystem. Magnetic excitations in Gd subsystem at $\nu \sim 3\,cm^{-1}$ for $T < 7\,K$ were revealed in addition to the high frequency Mn AFMR mode at $\nu > 20\,cm^{-1}$.

Thus we observed a complex character of the Mn ordering with decreasing of the ionic radii from Sm to Gd, a significant contribution of Sm and Gd ions to the low temperature magnetic properties and new rare-earth branches of magnetic excitations in the manganites studied.

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