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Electric polarization induced by phase separation domains in multiferroics of RMn_2O_5 (R = Gd, Bi)

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A comparative study of the electric polar states in multiferroics $GdMn_2O_5$ and $BiMn_2O_5$ with a common subsystem of Mn ions (in equal ratios of Mn^{3+} and Mn^{4+} ions), but strongly differing in their properties of R ions, was carried out. Ion Gd^{3+} (ground state $^8S_{7/2}$), which is a strongly magnetic ion with spin S = 7/2, but weakly interacting with the lattice. Ion Bi^{3+} is a non-magnetic ion containing $6s^2$ electrons, which cause a strong non-central local distortion of the lattice. The long-range ferroelectric order having an exchange-striction magnetic nature had been observed at low temperatures ($T \leq T_C$ = 30-35 K).

The homogeneous single-domain $GdMn_2O_5$ the maximal ferroelectric polarization, as compare with other RMn_2O_5, was observed due to the strong homogeneous Gd-Mn exchange. In $BiMn_2O_5$, the lattice distortions near the Bi ions changed the distances between pairs of ions Mn of different valences, violating the homogeneity of the internal field of the exchange striction. As a result, the polarization induced by the exchange striction, was in 20 times less. Along with this, an electrical polarization of a different nature had been found at temperatures $T >> T_C$ in both studied crystals [1]. A generally accepted point of view is that RMn_2O_5 has sp.gr. *Pbam* (at room temperature) not admitting the existence of a polar order. We believe that the high-temperature polarization we observed was caused by the frozen superparaelectric state, which was formed by the restricted polar domains resulting from phase separation and charge carrier self-organization. Charge ordering in RMn_2O_5 and the e_g electron transfer between Mn^{3+}–Mn^{4+} ion pairs are key factors responsible for polar electric states of these multiferroics at all temperatures. The double exchange between Mn^{3+}-Mn^{4+} ions induces the phase separation in RMn_2O_5 which is similar to phase separation in $LnAMnO_3$ (A = Sr, Ba, Ca) manganites containing Mn^{3+} and Mn^{4+} ions as well. Phase separation exists at all temperatures and makes the formation of local conductive domains containing Mn^{3+}-Mn^{4+} ion pairs with ferromagnetically oriented spins energetically favorable. The polar phase separation domains are located in a dielectric antiferromagnetic (paramagnetic) matrix of the original crystal, forming at sufficiently low temperatures the frozen superparaelectric state. Such type state was considered theoretically in the system of isolated ferroelectric nanoscale domains in a dielectric matrix [2]. In this state, hysteresis loops and remanent polarizations are observed. The frozen superparaelectric state turns into the conventional superparaelectric one near T^* (T^* = 100-330 K), in which the hysteresis loops are destroyed. The T^* values correspond to the temperatures at which the potential barriers of the restricted polar domain reorientations become equal to the kinetic energy of the itinerant electrons (leakage). The polarization magnitudes, its anisotropy, and the temperatures T^* were very different for $GdMn_2O_5$ and $BiMn_2O_5$. The magnetic field H increased the barriers at the phase separation domain boundaries due to the double exchange growth, thus increasing the T^*. The electric hysteresis loops were measured using the version of the PUND method presented in [3], which was adapted to studies of the local polar domains. The emergence of the local phase separation domains was confirmed in the high-sensitivity 3-crystal X-ray diffractometer measurements and in the permittivity and conductivity investigations. We revealed correlations between properties of the local phase separation domains and hysteresis loops.

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