

# Magneto-electric coupling in granular multiferroics

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Currently composite materials with combined magnetic and electric degrees of freedom attract much of attention for their promise to produce new effects and functionalities<sup>1,2</sup>. The idea of using ferromagnetic and ferroelectric properties in a single phase multiferroics was developing since seventeenth. However, in bulk homogeneous materials this coupling is weak due to relativistic parameter  $v/c$ , with  $v$  and  $c$  being the electron velocity and the speed of light, respectively. Only recently the new classes of two-phase multiferroic materials such as single domain multiferroic nanoparticles<sup>3</sup>, laminates<sup>4</sup>, and epitaxial multilayers<sup>5</sup> and granular materials<sup>6</sup> were discovered giving a new lease of life to this field. So far, the interface strain generated by the ferroelectric layer was considered as the promising mechanism for strong enough magneto-electric coupling in two-phase multiferroic materials. This strain modifies the magnetization in the magnetic layer and the magnetic anisotropy energy.

We propose a different mechanism for magneto-electric coupling emerging at the edge of strong long-range electron interaction, ferroelectricity, and magnetism<sup>7</sup>. In composite multiferroics - materials consisting of metallic ferromagnetic grains embedded into ferroelectric (FE) matrix, the origin of this coupling is twofold: i) Strong influence of FE matrix on the Coulomb gap depending the electron localization length and the overlap of electron wave functions, and therefore controlling the exchange forces. ii) Dependence of the long-range part of Coulomb interaction, and thus the exchange interaction, on the dielectric permittivity of the FE matrix.

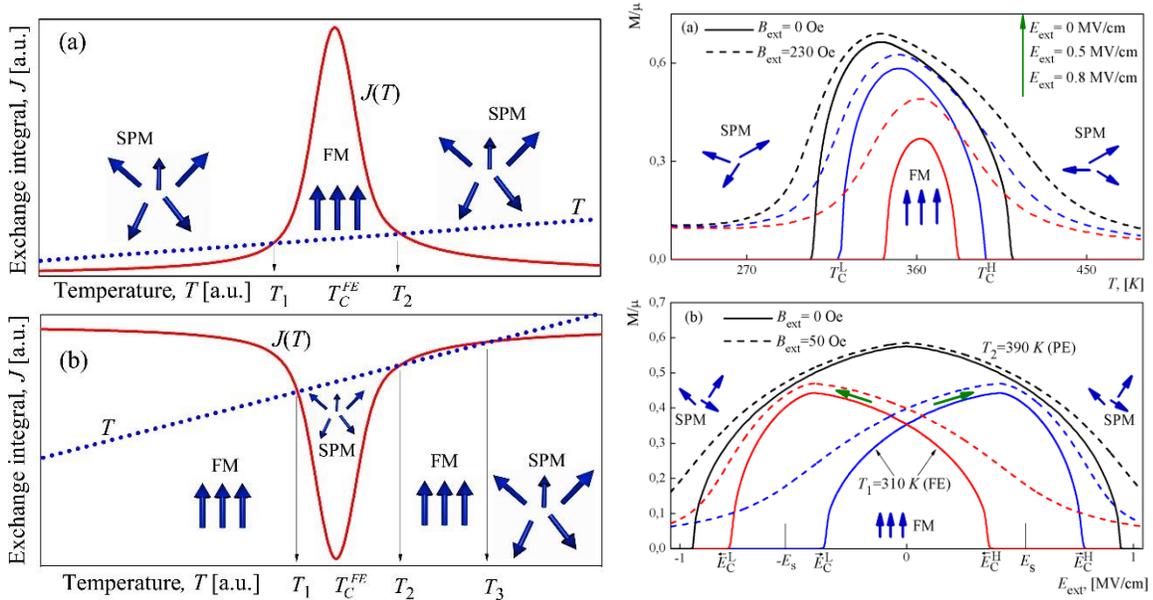


Figure 1. Left panel is the dependence of intergrain exchange constant in granular multiferroic on temperature. (a) and (b) correspond to granular systems with different parameters. Dotted line shows temperature.  $T_C^{FE}$  is the FE Curie temperature. Right panel shows magnetization of granular multiferroic as a function of temperature (a) and external electric field (b). SPM denotes superparamagnetic state of the multiferroic, FM denotes ferromagnetic state.  $B_{ext}$  and  $E_{ext}$  are external magnetic and electric field, correspondingly.

We show that the effective ferromagnetic exchange constant  $J$  between the ferromagnetic grains strongly depends on temperature near the ferroelectric Curie temperature in granular multiferroics due to the above mentioned mechanisms. This give rise to unusual magnetic phase diagram of composite multiferroics (see Fig. 1, left panel). The transition temperature between ordered and disordered magnetic states can be found approximately using the equation  $J(T) = T$ . FM state corresponds to  $J(T) > T$ . If mechanism (i) is the strongest, the FM state appears at higher temperatures than the disordered SPM state. This is known as an inverse phase transition.

We study magnetization of composite multiferroic as a function of temperature and external electric field (see Fig. 1, right panel). The FM state exists at finite temperature range around the FE phase transition point. Outside this region the superparamagnetic phase appears. Both the magnetic phase transition temperature and the magnitude of magnetization are strongly electric field dependent. In addition, the magnetic phase transition can be drive by the external electric field. The magnetic state of the system depends on the mutual orientation of external electric field and polarization of FE matrix. The ferromagnetic and ferroelectric degrees of freedom are coupled due to the influence of FE substrate on the screening of intragrain and intergrain Coulomb interaction.

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