Colossal magneto-capacitive coupling in multi-ferroic spinel chalcogenides

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The normal cubic spinel systems $AB_2X_4$ ($X$=S,Se) exhibit a wide range of exceptional ground state properties depending on the appropriate choice of $A$- and $B$-site ions. In the case of CdCr$_2$S$_4$ the Cd$^{2+}$-ions on the $A$-sites are non-magnetic due to completely filled 4$d$-shells. The electronic configuration of the Cr$^{3+}$ ions on the octahedral coordinated $B$-sites is determined by a 3-fold occupation of the low-lying $t_{2g}$-levels resulting in a total spin of $S = 3/2$. The magnetic coupling of the $B$-site sublattice leads to a ferromagnetic transition below $T_c = 84$ K. At the same time the system is not Jahn-Teller active. The absence of an orbital degree of freedom enables the structural degeneracy to be lifted by local dipolar distortions. As a consequence multi-ferroic behavior, namely the coexistence of ferromagnetism and relaxor ferroelectricity can be detected. In addition, the onset of spontaneous magnetization strongly influences the relaxor dynamics which leads to a very pronounced magneto-capacitive effect of up to 500% close to $T_c$.


This work was partly supported by the Bundesministerium für Bildung und Forschung via grant No. VDI/EKM 13N6917-A and by the Deutsche Forschungsgemeinschaft (SFB 484)