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Ferroelectric magnets: a Conical Spiral and an Ising Chain

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Multiferroics, where magnetic order with broken inversion symmetry accompanies the occurrence of ferroelectric polarization, can show remarkable tunability of dielectric properties by applied magnetic fields, such as reversibly flipping of ferroelectric polarization or a drastic change of dielectric constant with fields. It turns out that the origin of ferroelectricity in most of new multiferroics is spiral magnetism that tends to produce uniform lattice distortions, i.e., ferroelectric distortions, through antisymmetric exchange coupling. Among such materials, spinel $CoCr_2O_4$, exhibiting a conical-spiral spin order, is unique in that it exhibits spontaneous magnetization as well as electric polarization. We have studied the detailed switching behavior of magnetoelectric domains, characterized by polarization, magnetization, and magnetic wave vector, under variation of temperature and applied magnetic fields. New aspects that we have discovered from this study include [1] polarization reversal at the magnetic lock-in transition temperature (T_L) with thermal variation or with isothermal variation of magnetic fields (without changing their direction) at a temperature near T_L, [2] surprisingly, this polarization reversal happens without change in spin rotation direction in spiral spins. We also present newly-discovered multiferroicity in a "collinear" chain magnet of $Ca_3(Co,Mn)_2O_6$. In the Ising chain magnet, "symmetric" exchange coupling seems responsible for the multiferroicity. We show that in $Ca_3Co_{2-x}Mn_xO_6$ ($x \approx 1$), the ferroelectricity originates from the coexistence of the alternating order of magnetic ions with different charges and the collinear up-up-down-down spin order in the frustrated spin chain. The system also exhibits magnetic freezing transitions that affect the temperature dependence of the electric polarization magnitude.