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Colossal magnetocapacitive effects in geometrically frustrated chalcogenide spinels

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It is well known that the spinel structure is susceptible to the occurrence of geometrical frustration, which in the past was invoked to explain a number of unusual observations concerning the magnetic and orbital degrees of freedom in these materials. We demonstrate that several chalcogenide spinels also exhibit very unusual dielectric behavior, especially an extremely strong coupling of magnetic and dielectric properties and the simultaneous occurrence of magnetic and polar order. Especially, in $CdCr_2S_4$ a colossal magnetocapacitive effect is observed, which shows up as a sharp upturn of the dielectric constant ε' when the sample becomes ferromagnetic and as a variation of ε' up to a factor of 30 when the sample is subjected to external magnetic fields. As revealed by linear and non-linear dielectric measurements, this material shows the typical signatures of relaxor ferroelectrics, i.e. a strong increase of the static dielectric constant with decreasing temperature and considerable frequency dispersion of the complex permittivity. While in most relaxor ferroelectrics the freezing of polar moments is driven by frustrated interactions related to substitutional disorder, in the present pure system geometrical frustration seems a plausible mechanism to explain the relaxor behavior. However, one may also speculate on completely different mechanisms of ferroelectric polarization, e.g., the ordering of electronic degrees of freedom. The concomitant occurrence of polar and magnetic order makes $CdCr_2S_4$ another example of the rare species of multiferroic materials. In contrast to other members of this group of materials, it has sizable ordering temperatures and moments. A detailed investigation of the relaxational dynamics in this material provides clear evidence that the observed magnetocapacitive effect stems from an enormous acceleration of the relaxation dynamics induced by the development of magnetic order. In addition, recent results reveal even larger magnetocapacitive effects in In-doped $CdCr_2S_4$. In addition to $CdCr_2S_4$, we found similar effects also in ferromagnetic $CdCr_2Se_4$ and, most astonishing, in $HgCr_2S_4$, which exhibits a complex type of antiferromagnetic magnetic order at low temperatures. In the latter system, the magnetocapacitive effect, exemplified by the relative increase of ε' in a field of 5 T, reaches values up to 8×10^5 %.