Novel incommensurate magnetic phase in the magnetoelectric Sr-doped cobaltate $CaBaCo_4O_7$

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The magnetetoelectric $CaBaCo_4O_7$ compound offers an interesting scenario to study frustrated magnetic configurations. The Co^{2+} and Co^{3+} ions in tetrahedral oxygen coordination form a three-dimensional framework of interconnected triangular and kagome layered arrangements [1]. The compound becomes ferrimagnetic below 60 K, and displays a strong increase of electric polarization of 17 000 $\mu C/cm^2$, driven by exchange-striction. In this work, we present our results on the thermal evolution of magnetic and crystallographic properties of powder samples of $Ca_{1-x}Sr_xBaCo_4O_7$ (x = 0, 0.02, 0.05, 0.07) to study the effect of substitution at the Ca site. We will show that low doping levels (<10 at.%) change quite dramatically the magnetic behavior of the compound, as observed in magnetization vs. temperature measurements. Combined with extensive use of Neutron Power Diffraction we analysed the evolution of the magnetic order as a function of temperature and composition of the samples. The reported non collinear ferrimagnetic order of the parent compound is only retained for the lowest doping level $x = 0.02$ and is accompanied by a strong unit cell distortion. In turn, further Sr doping blurs this distortion and favors other magnetic arrangements. In the temperature range 62 K < T < 82 K, samples with $x \geq 0.02$ show a plateau in the magnetization. By using the superspace group theory and its implementation in the Rietveld refinement of neutron diffraction data, we have solved the incommensurate magnetic structure that appears at these intermediate temperatures. The magnetic order has a propagation vector k = (1/2, 1/2, g) with $g \approx 0.02$ and it belongs to the superspace group $Pna2_11'(1/2, 1/2, g)qq0s$. This phase corresponds to a modulated spin structure with distinct behaviors of the triangular and kagome cobalt sites and could explain previous findings reported in the literature for other substitution sites in the $CaBaCo_4O_7$ family.

[1] V. Caignaert, V. Pralong, A. Maignan, B. Raveau. Solid State Communications 149 ,453 (2009)

Keywords: Magnetism, Frustated, Cobaltate, magnetetoelectric, incommensurate order