Increase of Magnetic Transition Temperatures by Reduction of Local Disorder for Perovskite Manganites

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ABSTRACT

We report the synthesis of $Sr_{1-x}Ca_xMnO_3$ and $La_{0.5}Ba_{0.5}MnO_3$ perovskites over extended cation and oxygen composition ranges and describe the dependence of their phase stability on the tolerance factor $t = t(x,T,\delta)$ that is a function of composition, temperature, and oxygen content. We show that magnetic transition temperatures depend strongly on the tolerance factor and charge disorder while dependence on the structural disorder is less important. By reducing charge and structural disorder we have significantly increased the Curie and Neel temperatures for perovskite manganites.

Perovskite manganites, $AMnO_3$, have been recently studied in great detail because of very interesting magnetic and electronic properties resulting from competing charge, exchange, and phonon interactions [1]. The properties can be tuned over a wide range through the choice of the sizes and charges of the A-site cations that control the degree of structural distortions and the formal valance of Mn. The correlation of physical properties with the Mn–O–Mn bond-angle controlled electronic band-width has been clearly established for decades [2,3]. These correlations are also frequently described in terms of the average structure using a concept of tolerance factor $t = [A\text{-}O]/\sqrt{2}[Mn\text{-}O]$ [4]. Here, [A-O] and [Mn-O] are the average cation-oxygen interatomic distances of the A- and B-sites, respectively, the approximate values of which can be obtained from the ionic sizes that are tabulated for various coordination numbers and oxidation states at room temperature [5]. It was reported recently that the ferromagnetic transition temperatures can be better accounted for by using in addition to the tolerance factor the variance of sizes of the A-site ions, $s(x) = \sqrt{\Sigma_i\{x_i[A_i\text{-}O]^2 - [A\text{-}O]^2\}}$, where $[A\text{-}O] = \Sigma_i x_i[A_i\text{-}O]$,