

## Response to the reviewer's remarks

### 1. There appears to be a missing pattern in Figure 10.

Thanks to the reviewer for this remark. The correct figure captions read:

Fig. 8. Real part  $M'$  and imaginary part  $M''$  of ac differential magnetization of S2 obtained by sonochemistry vs temperature at  $H=10$  Oe,  $f=1$  kHz [28]

Fig. 10. The differential ac susceptibility of S2 obtained by sol-gel auto-combustion in ac magnetic field of 10 Oe and frequency of 1 kHz.

### 2. Could we provide a detailed explanation to validate if the additional peaks observed in the Neutron Diffraction (ND) stem from the helical spin arrangement?

The necessary information is given in the amended version.

### 3. Upon inspecting the SEM images, it's clear that the particles in Sample S2 is significantly larger than in S1. Could the disparity in magnetic properties between S1 and S2, such as critical temperature, be attributable to size or boundary effects as commonly seen in nanostructured materials?

Figure 7 and Figure 9 display the change of magnetization with temperature in dc and ac magnetic field measurements for sample S1 obtained by the two methods - auto-combustion and sonochemistry. The S1 sample obtained by auto-combustion consists of particles of a very non-uniform shape whereas the S1 particles obtained by sonochemistry are with hexagonal shape. As can be seen from Fig.7 and Fig.9 the differences in the particle shape and size did not affect appreciably the critical temperature of different magnetic transitions. Only for the sample S1 obtained by sonochemistry with particles of hexagonal shape, the transitions are better expressed and the metamagnetic one can be detected. We believe that this is associated with the well-built hexagonal shape characteristic of hexaferrites.

In our previous studies on the effect of particle shape on the magnetic properties of M-type hexaferrites, we found that for size to have a significant effect on the magnetic properties, the thickness of the hexagonal particle must be below 50 nm or the particle to has a shape close to spherical and a size below 120 nm far below the critical size for single domain state. In our case, the particles are polydomain and their thickness above 100 nm seems sufficient so that not to feel the influence of surface defects (through an essential imbalance of the long-range spin arrangement) on the magnetic phase transitions.

### 4. If the variation in magnetic behavior is triggered by intrinsic distortion due to Aluminium substitution, what level of distortion can induce a shift in the critical temperature or potentially modify the magnetic phase?

The magnetic phase transitions in  $Al_x$ -BSZFO (*Al-doped BSZFO*) as a function of Al concentration below 400 K has been the subject of the article by Chang, Lee et al. "Al doping effect on magnetic phase transitions of magnetoelectric hexaferrite  $Ba_{0.7}Sr_{1.3}Zn_2(Fe_{1-x}Al_x)_{12}O_{22}$ ", Phys. Rev. B 85, 064402(2012).

Based on using single-crystal neutron diffraction and magnetization measurements, this cited study sheds light on the physical picture that: " *$Al^{3+}$  doping leads to a reduction of the easy-plane anisotropy and hence to the appearance of axial spin components along the c axis, and subsequently stabilizes longitudinal conical (LC) spin structures. Then, the finite magnetoelectric polarization can easily appear when the LC configuration flops into a transverse orientation under small external magnetic field.*"

These authors claim that "*the doping dependence of the magnetoelectric property is closely tied to the evolution of magnetic structure*" and classify Al-doped BSZFO into three regimes: *underdoped* for  $x \leq 0.04$ , *optimally doped* for  $0.04 < x \leq 0.08$ , and *overdoped* for  $0.08 < x$ .

Their final conclusion reads: "*Al doping stabilized longitudinal conical phases up to  $x < 0.10$ , beyond which commensurate ordering took place.*"