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## ARTICLE

# Synthesis and characterization of the new two-dimensional Heisenberg antiferromagnet double perovskite BaLaCuSbO<sub>6</sub>

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BaLaCuSbO<sub>6</sub> double perovskite has been successfully synthesized by solid state reaction in air atmosphere. Its structure was refined using powder neutron diffraction in the monoclinic space group I 2/m with a 4% *antisite* disorder on the B cations. Magnetic measurements give signs of 2D-antiferromagnetic behaviour with T<sub>N</sub> around 64 K. The Jahn-Teller distortion produced by Cu<sup>2+</sup> ions favours a crystallographic tetragonal distortion and consequently the in-plane super superexchange antiferromagnetic interactions, J<sup>90°</sup>, are favoured over the in-plane J<sup>180°</sup> antiferromagnetic exchange interaction. Both, J and J' magnetic interactions have been evaluated according to a Heisenberg antiferromagnetic rectangular model using an approximation to Curie's law in powers of J/T, being |J| around 10 times stronger than |J'|.

## Introduction

In ordered double perovskites (DP) with general formula AA'BB'O<sub>6</sub>, B and B' ions are distributed in the octahedral sites in a rock salt like arrangement by sharing corners. Both A and A' ions occupy the cuboctahedral sites formed by this network of octahedra.

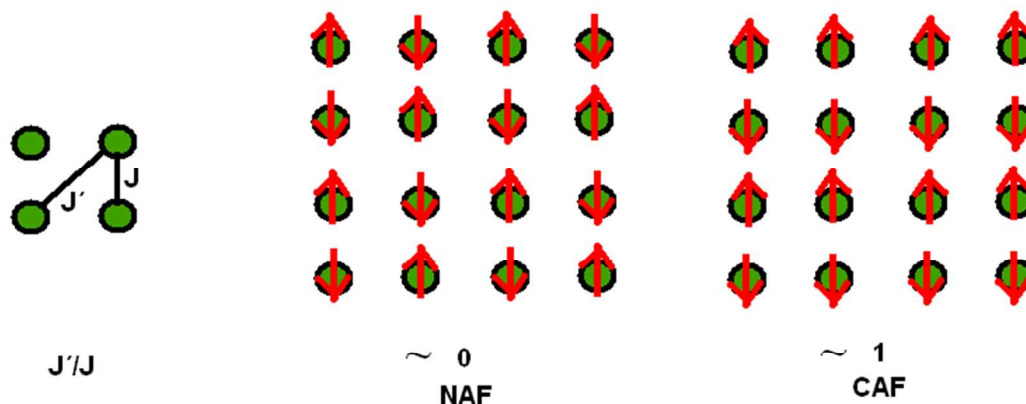
DP in which B-site is occupied by a magnetic transition metal ion, and B'-site is occupied by a nonmagnetic one have been extensively studied [1-4]. These DP oxides are of particular interest because of their different magnetic and electrical properties, which are strongly altered by the nature and oxidation state of the transition metal ion. Particularly, several Cu<sup>2+</sup> containing DP of the type AA'CuB'O<sub>6</sub> have already been studied. The structures of SrLaCuRuO<sub>6</sub> and SrLaCuSbO<sub>6</sub> were reported by Attfield *et al* [5], however no magnetic studies were informed. Several A<sub>2</sub>CuB'O<sub>6</sub> were studied by Iwanaga *et al.* [6] and cooperative Jahn-Teller (JT) effect and anomalous antiferromagnetic behaviour was reported.

Since Cu<sup>2+</sup> ion in a regular octahedral environment has an electronic configuration t<sub>2g</sub><sup>6</sup>e<sub>g</sub><sup>3</sup>, its ground electronic configuration is orbitally degenerated and asymmetrically filled, then in order to remove the degeneracy and lower the energy, the JT distortion is produced, which is usually expressed as the elongation of the octahedra along the z axes.

Regarding the g values, and considering only oxides, there is a wide range of g values which goes from 1.9386 (for Cu<sub>0.96</sub>Mn<sub>0.04</sub>GeO<sub>3</sub>, [7]) to 2.126 (for Y<sub>2</sub>BaCuO<sub>5</sub>, [8]). Since we didn't make experiments to calculate g, we actually do not know the exact value of g for our compound.

Antiferromagnetic DP with B-Cation order, of the type AA'BB'O<sub>6</sub> (where B is a paramagnetic ion and B' a diamagnetic one) show significant geometrical magnetic frustration (GMF) [1]. The origin of this GMF is because the magnetic ions are located at the corners of a regular tetrahedron. On the other hand, this GMF is absent, or substantially decreased, when DP have cooperative JT distortion because the distances between the magnetic ions, located in the corners of this tetrahedron are not equivalent [1].

**Figure 1:** a) Square lattice formed only by magnetic ions laying on a plane and corresponding to magnetic couplings  $J$  and  $J'$ , b) Neel Antiferromagnetic (NAF) Square Lattice, c) Collinear Antiferromagnetic (CAF) Square Lattice.



Owing to cooperative JT distortion in  $AA'B'O_6$  DP the bond lengths of the  $\text{CuO}_6$  octahedra in the  $xy$  planes are  $\sim 2 \text{ \AA}$  and  $\sim 2.3 \text{ \AA}$  along the  $z$  axes, on the other hand the  $B'O_6$  octahedra (with the non-magnetic ion  $B'$  in the centre) are almost regular, although with a slight compression along the  $z$  axes [9]. Thus with the  $\text{Cu}^{2+}$   $d_{xz}$ ,  $d_{yz}$  and  $d_z^2$  orbitals filled and the  $d_{x^2-y^2}$  half filled, there is no magnetic interactions along the  $z$  axes and this makes the magnetic interactions two-dimensional (2D) [9]. Thus, this constitutes a magnetic square lattice with two magnetic coupling constants,  $J$  for the nearest-neighbour interaction and  $J'$  for the next nearest-neighbour interaction (see left side in Figure 1).

Considering low dimensional materials, there is a difference between a two-dimensional and a quasi-two dimensional magnetic arrangement. For a quasi-two dimensional compound, there are still magnetic couplings between successive planes of square lattices, and the inter-plane magnetic interactions are usually lower than the intra-plane ones. In an isolated two-dimensional magnetic material, there are no magnetic coupling between planes [10]. In a two dimensional antiferromagnetic compound the presence of magnetic frustration will depend on the character of the magnetic interactions competing in the compound [11]. According to the ratio  $|J'/J|$ , both a two-dimensional and a quasi-two dimensional system with antiferromagnetic interactions can be either a Neel Antiferromagnet (NAF) if  $|J'/J| \leq 0.4$  or a Collinear Antiferromagnet (CAF) if  $|J'/J| \geq 0.7$  (see central and right panels in Figure 1) [9]. When both magnetic interactions,  $J$  and  $J'$ , are antiferromagnetic ones, then there is frustration involved, independently of the final magnetic order (NAF or CAF) for such system. This frustration is larger when  $J$  and  $J'$  are similar. Since weak inter-plane interactions can be present, two-dimensional to three-dimensional magnetic crossover is naturally expected to occur at low temperatures [12, 13].

A DP, which gets together a rock salt arrangement of a  $d^9$  active JT ion like  $\text{Cu}^{2+}$  and an empty  $p$  orbitals  $d^{10}$  ion like  $\text{Sb}^{5+}$ , can be a good candidate to study low dimensional magnetic

behaviour. Thus, we synthesized the new DP  $\text{BaLaCuSbO}_6$ , which has  $\text{Cu}^{2+}$  and  $\text{Sb}^{5+}$  ions distributed over two crystallographic distinguishable sites where  $\text{Cu}^{2+}$  presents a strong JT distortion.

In the present paper we performed the structural characterization of  $\text{BaLaCuSbO}_6$  with RT PXRD and NPD and the corresponding magnetic characterization. The later allow us to explore the low dimensional magnetic behaviour and measure the magnetic interactions. For this purpose, we use an analytical function that fits the susceptibility curves for a 2D spin- $1/2$  quantum Heisenberg antiferromagnet (QHAF). This function has been formulated for the Heisenberg AF rectangular model using a modified Padé rational approximation to Curie's law in powers of  $J/T$ . For the best of our knowledge, this is the first time that both,  $J$  and  $J'$  have been quantified in a DP oxide.

## Experimental section

$\text{BaLaCuSbO}_6$  was synthesized by solid-state method from the starting materials, weighed in stoichiometric amounts:  $\text{BaCO}_3$ ,  $\text{La}_2\text{O}_3$ ,  $\text{CuO}$  and  $\text{Sb}_2\text{O}_3$ , all in analytical grade. The sample was heat-treated at  $750 \text{ }^\circ\text{C}$ ,  $850 \text{ }^\circ\text{C}$ , and  $1100 \text{ }^\circ\text{C}$ , in the three cases for 12 h in air atmosphere, and with regrindings during 15 min between heat-treatments. Heating and cooling rates were  $3^\circ \text{C/min}$ .

Powder X-ray diffraction (PXRD) pattern was measured on a PANalytical X'Pert Pro diffractometer (40 kV, 40 mA) in Bragg-Brentano geometry with  $\text{Cu K}\alpha$  radiation at room temperature, between  $5^\circ$  and  $120^\circ 2\theta$  in steps of  $0.02^\circ$  and counting time of 7 sec. Powder neutron diffraction (PND) pattern was collected in the D1A powder diffractometer at Institute Laue-Langevin, Grenoble, France, at room temperature (RT). The measurement was performed between  $0^\circ$  and  $128^\circ$  in  $2\theta$  with steps of  $0.1^\circ$ , and the wavelength used was  $1.91 \text{ \AA}$ . Refinements were performed by the Rietveld method [14] using the Fullprof program [15]. A pseudo-Voigt shape function was always adequate to obtain good fits.

The magnetic characterization was performed in a commercial SQUID magnetometer on a powdered sample, between 5 and 300 K. The magnetization ( $M$ ) vs. temperature ( $T$ ) curves were measured under Zero Field Cooling (ZFC) and Field Cooling (FC) conditions under three different applied fields, of 0.5, 1.0 and 5.0 T.

## Results

### Structural characterization

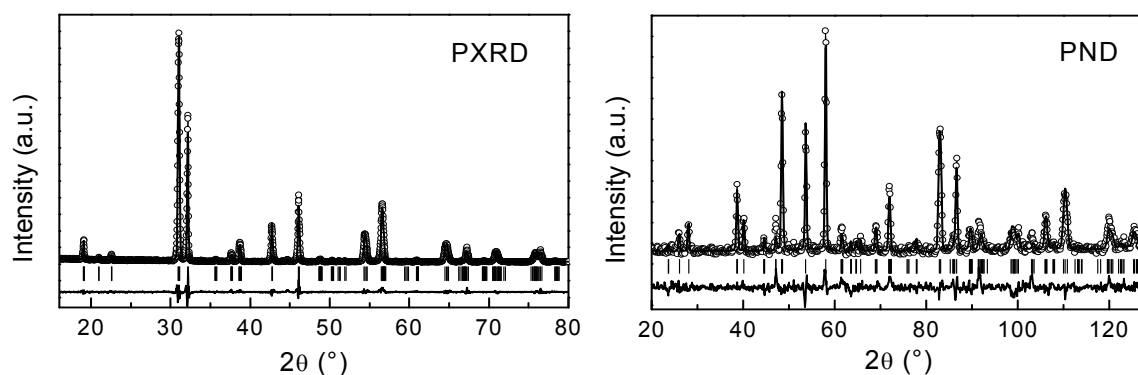
A pure BaLaCuSbO<sub>6</sub> DP was obtained as a dark greenish powder. The determination of the space group (SG) was made in the following way: by autoindexing the PXRD data we determine that the Crystal System was monoclinic. Specifically, in the case of ordered double perovskites, the article by P. Woodward and co-workers [16], classifies ordered double perovskites in the allowed SG's based on the tilt of octahedra and ordering of B cations, and the only possible monoclinic SG's are C2/c, P2<sub>1</sub>/c and C2/m (I2/m with another origin choice). Using high quality laboratory PXRD data we found the best possible SG for this compounds was I2/m. **Figure 2**, left side, shows the RT-PXRD pattern and corresponding Rietveld refinement in the I2/m space group. **Figure 2**, right side, shows the refinement of the RT-NPD data, which was jointly performed with the RT-PXRD refinement. No impurities were detected. Refined cell parameters are listed in **Table 1**, upper panel. The tetragonal distortion  $c/(\sqrt{2}a) = 1.07$  is in agreement with other Cu<sup>2+</sup> containing DP showing typical cooperative JT distortion [9, 17]. The same DP, BaLaCuSbO<sub>6</sub>, is informed in

the Powder Diffraction Files Database, indexed in the P 2<sub>1</sub>/n space group, PDF-number 047-0279 [18], but no additional characterization has been reported at present time.

**Table 1** lists the atomic coordinates, isotropic thermal factors and occupancies for all ions. This refined monoclinic structure has two octahedral sites,  $2d$  and  $2a$ , with an ordered distribution of Cu<sup>2+</sup> and Sb<sup>5+</sup> ions, in the classical rock salt like arrangement, with an *antisite* disorder of about 4%. The  $2d$  Wyckoff site contains almost all Cu<sup>2+</sup> ions. The oxygen occupancies were also refined but they remained close to one within the standard deviations so they were fixed to unity. These results confirmed the proposed stoichiometry BaLaCuSbO<sub>6</sub>.

**Table 1** also shows main distances and angles for BO<sub>6</sub> and B'O<sub>6</sub> octahedra. The distances corresponding to  $2d$  site, mainly occupied by Cu<sup>2+</sup> ions, show noticeable differences between the two distances associated to  $c$  axes and the four distances associated to  $ab$  plane, because of the strong JT distortion. The  $2a$  site, almost totally occupied by Sb<sup>5+</sup> ions, shows small differences between the different axes although there is a little compression along the  $c$  axes. The tilt system  $a^-b^-c^0$ , associated to the space group I 2/m is illustrated in **Figure 3**. On the left side no tilt is observed along the  $c$  axes. On the right side *antifase* tilt, along  $a$  and  $b$  axes of the pseudocubic cell is shown. The obtained tilt angles (**Table 1**) are small and the structure is composed by  $ab$  planes where distances between neighbour Cu<sup>2+</sup> ions inside a plane are smaller than between consecutive planes. The JT distortion is illustrated in **Figure 3**, right side: the dark grey octahedra (green online) are elongated along  $c$  axes.

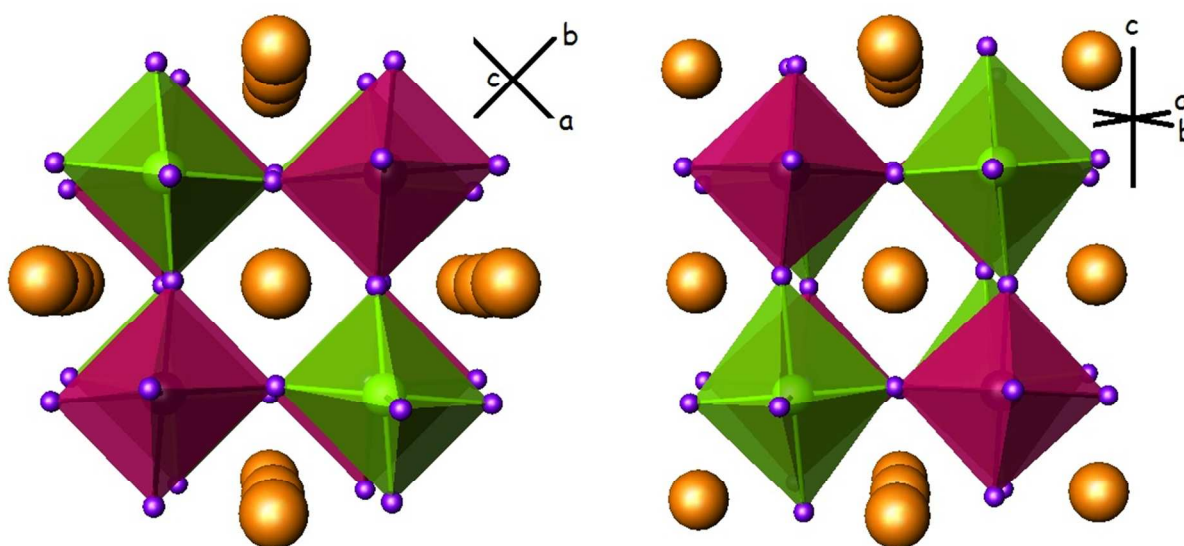
**Figure 2:** Rietveld refinement of PXRD and PND data at RT for BaLaCuSbO<sub>6</sub>. Observed (circles), calculated (line), and difference (bottom line), Vertical bars represent positions of Bragg reflections, space group I 2/m. Reliability factors: PXRD:  $\chi^2 = 2.46$ , Rwp = 14.7, Rp = 14.6, Bragg R-factor = 3.07, and: PND:  $\chi^2 = 1.13$ , Rwp = 29.4, Rp = 33.0, Bragg R-factor = 10.5.



**Table 1:** Lattice parameters, cell volume, atomic positions, thermal isotropic factors, bond distances and selected angles as determined by joint Rietveld refinements of RT PXRD and PND for BaLaCuSbO<sub>6</sub>.

Cell	a (Å)	b (Å)	c (Å)	β (°)	V (Å <sup>3</sup> )	Space group	
	5.5683(5)	5.5683(4)	8.4637(3)	90.288(1)	262.42(3)	I 2/m (#12)	
T (K)	atom	Site	x	y	z	B <sub>iso</sub>	Occupation
300	Ba/La	4i	0.4965(1)	0	0.2520(1)	0.353(3)	0.5/0.5
	Cu/Sb(1)	2d	0	0	1/2	0.65(2)	0.956(2)/0.044(2)
	Cu/Sb(2)	2a	0	0	0	0.09(1)	0.044(2)/0.956(2)
	O1	4i	-0.0420(3)	0	0.2352(2)	1.15(5)	1
	O2	8j	0.2351(2)	0.2741(2)	0.0136(2)	1.77(2)	1
Distances (Å)	(B – O1) <sub>2d</sub>	2.252(2)x2	B' – O1	2.006(2)x2			
	(B – O2) <sub>2a</sub>	1.942(1)x4	B' – O2	2.014(1)x4			
	< B – O >	2.045	< B' – O >	2.011			
Angles (°)	B – O1 – B'	167.3(1)	δ	6.4			
	B – O2 – B'	168.8(1)	δ	5.6			

**Figure 3:** Octahedra Tilt and JT distortion; left: in *ab* plane, right: along *c* axes. White spheres: Ba/La, black spheres: O, dark grey octahedra (green on line): CuO<sub>6</sub>, light grey octahedra (pink on line): SbO<sub>6</sub>.



### Magnetic characterization

M vs. T measurements were done under three different applied fields (H), 0.5, 1.0 and 5.0 T, under ZFC and FC conditions (see **Figure 4**). Fits of the high temperature region ( $200 < T < 300$  K) with Curie-Weiss law, according to  $\chi = C/(T - \theta)$  are shown in **Figure 4**, right side (red lines on line). C is the Curie constant,  $\theta$  is the Weiss temperature and  $\mu_{\text{eff}}$ , the effective magnetic moment calculated from the Curie constant (See **Table 2**). The presence of moderated strong antiferromagnetic interactions can be deduced from the negative sign of Weiss temperatures. The experimental effective magnetic moments

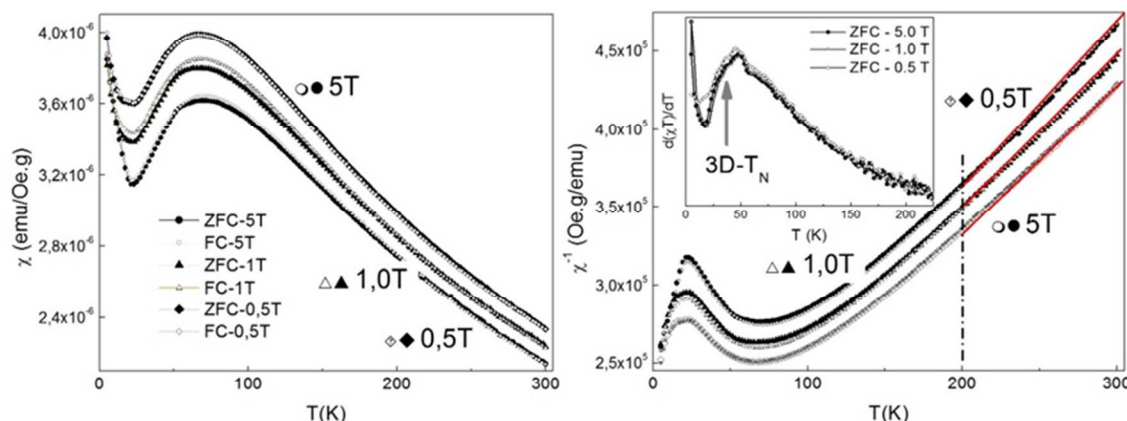
are higher than the theoretical ones corresponding to spin only contributions, suggesting some orbital contribution.

$\chi$  vs T data have a broad maximum about 70 K, and a minimum about 20 K. This broad maximum in susceptibility is compatible with 2D antiferromagnetic behaviour, which can be understood by considering the orientations of the *d* orbitals of Cu<sup>2+</sup>.

Low dimensional behaviour has been already reported for some Cu<sup>2+</sup> containing DP. Tetragonally distorted B-site ordered perovskites synthesized and characterized by Iwanaga, D. *et al.* [6] show two-dimensional antiferromagnetic behaviour because

of the in-plane superexchange interactions. The  $\text{Sr}_2\text{Cu}(\text{W}_{1-x}\text{Mo}_x)\text{O}_6$  DP, studied by Vasala, S. *et al.* [9] are other examples of quasi-two dimensional  $S = \frac{1}{2}$  square lattices, however no experimental determination of  $J$  and  $J'$  were performed.

**Figure 4:**  $\chi$  vs  $T$  at three applied fields, on the left, and Curie-Weiss linear fittings  $\chi^{-1}(T)$ , on the right (red on line). Inset:  $d\chi/dT$  vs.  $T$ .



**Table 2:** Curie Constants ( $C$ ), Weiss Temperatures ( $\theta$ ), Effective Magnetic Moments ( $\mu_{\text{eff}}$ ), 2D-Neel Temperatures ( $T_N^{2D}$ ), and 3D-Neel Temperatures ( $T_N^{3D}$ ) from Curie-Weiss linear fittings.

H (kOe)	C (emu.K/Oe.g)	$\theta$ (K)	$\mu_{\text{eff}}(\mu_B/\text{Cu}^{2+})$	2D- $T_N$ (K)	3D- $T_N$ (K)
5 - ZFC	$1.087 \cdot 10^{-3}$	-163	2.2	66	46
5 - FC	$1.011 \cdot 10^{-3}$	-166	2.1		
10 - ZFC	$1.045 \cdot 10^{-3}$	-165	2.2	66	46
10 - FC	$1.021 \cdot 10^{-3}$	-156	2.1		
50 - ZFC	$9.807 \cdot 10^{-3}$	-153	2.1	74	46
50 - FC	$9.500 \cdot 10^{-4}$	-144	2.1		
$\mu_{\text{theo}} = 1.73 \mu_B$ (Spin only); $\mu_{\text{theo}} = 3.55 \mu_B$ (Orbital contribution)					

A 2D-Neel temperature has been obtained from the broad maximum of magnetization curves (see **Table 2**). The estimated. 3D-Neel Temperature in **Table 2** has been obtained from the maximum of  $d\chi/dT$  curves, showed in the inset of **Figure 4**. [19, 20].

We consider that this square lattice of spin- $\frac{1}{2}$   $\text{Cu}^{2+}$  ions which interact with each other via two orthogonal superexchange pathways is a spin- $\frac{1}{2}$  QHAF. Keith *et al.* have reported an analytical function that fits the susceptibility curves simulated for a 2D QHAF [21]. This function has been formulated for the Heisenberg AF rectangular model using a modified Padé

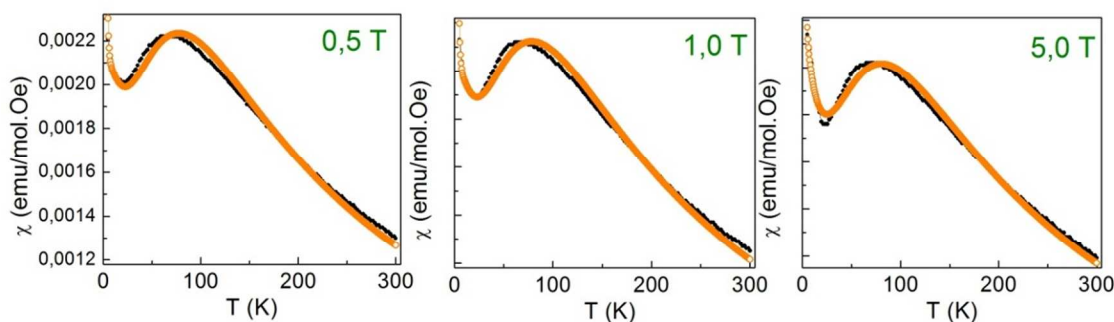
rational approximation to Curie's law in powers of  $J/T$ . The function for reduced susceptibility is:

$$\chi^*(\alpha, T) = \chi(\alpha, T)J / (Ng^2\mu_B^2) = (1/4T) (1 + \sum_{n=1}^6 N_n (J/T)^n) / (1 + \sum_{n=1}^6 D_n (J/T)^n)$$

Where  $\alpha = J'/J$ ,  $\mu_B$  is the Bohr magneton,  $N$  the number of spins,  $g$  the gyromagnetic ratio and  $N_n(x) = \sum_{m=0}^4 (N_{nm} x^m)$ ,  $D_n(x) = \sum_{m=0}^4 (D_{nm} x^m)$  [21]. Assuming a certain percentage  $\chi_p$  of residual paramagnetic contribution, it is possible to write (This analytical expression was provided to us by M. M. Turnbull):

$$\chi(\alpha, T) = (\chi_p/100)C/(4T) + (1 - \chi_p/100) C \chi^*(\alpha, T)$$

**Figure 5:** Non-linear fitting of  $\chi$  vs  $T$  curves using a modified Padé rational approximation to Curie's law in powers of  $J/T$ .



**Table 3:** Results obtained from polynomial fittings according to 2D Heisenberg Antiferromagnetic model,  $\chi_p$  is the paramagnetic susceptibility, C the Curie constant, and J and J' the magnetic coupling constants.

H (kOe)	$\chi_p$ (%)	C (emu.K/Oe.mol)	J (K)	$\alpha$	J' (K)
5 - ZFC	4.706	0.503	-127	0.086	-11
5 - FC	4.815	0.501	-126	0.087	-11
10 - ZFC	4.691	0.483	-128	0.085	-11
10 - FC	4.761	0.481	-126	0.086	-11
50 - ZFC	4.524	0.465	-131	0.057	-7
50 - FC	4.606	0.465	-131	0.057	-7

$\chi_p$  can come from the temperature independent paramagnetism of  $\text{Cu}^{2+}$  ion ( $60 \times 10^{-6}$  emu/mol) [20] and some crystal lattice defects or surface  $\text{Cu}^{2+}$  ion which are unpaired.

Using this analytical expression, non-linear fittings of our experimental data were performed along the whole temperature range by means of the least squares method. To achieve the minimization two different algorithms were used obtaining the same results: the Levenberg–Marquardt algorithm [23–24] as is implemented in the Gnuplot software [25] and the Nelder–Mead algorithm [26] implemented in a homemade fortran 90 code. **Figure 5** show the resulting fits, together with the  $\chi$  data for ZFC condition at the three magnetic fields. **Table 3** shows the obtained values for  $\chi_p$ , C, J and  $\alpha$  parameters, with the corresponding value for  $J' = \alpha J$ .

The presence of two significant antiferromagnetic interactions is confirmed by the negative signs of both, J and J'. The absolute value of J is one order of magnitude higher than the one of J', this indicates two quite different antiferromagnetic interactions and the possibility of some magnetic frustration.

Regarding magnetic interactions information obtained in literature, only the antiferromagnetic superexchange interactions, J, for DP oxides  $\text{Ba}_2\text{CuWO}_6$ ,  $\text{Sr}_2\text{CuWO}_6$ ,  $\text{Sr}_2\text{CuTeO}_6$  and  $\text{Ba}_2\text{CuTeO}_6$ , were evaluated by a high-temperature series expansion method, by Iwanaga, D. *et al.* [6], and the J values obtained have the same magnitudes than the J values determined in this work. On the other hand Vasala *et al.*

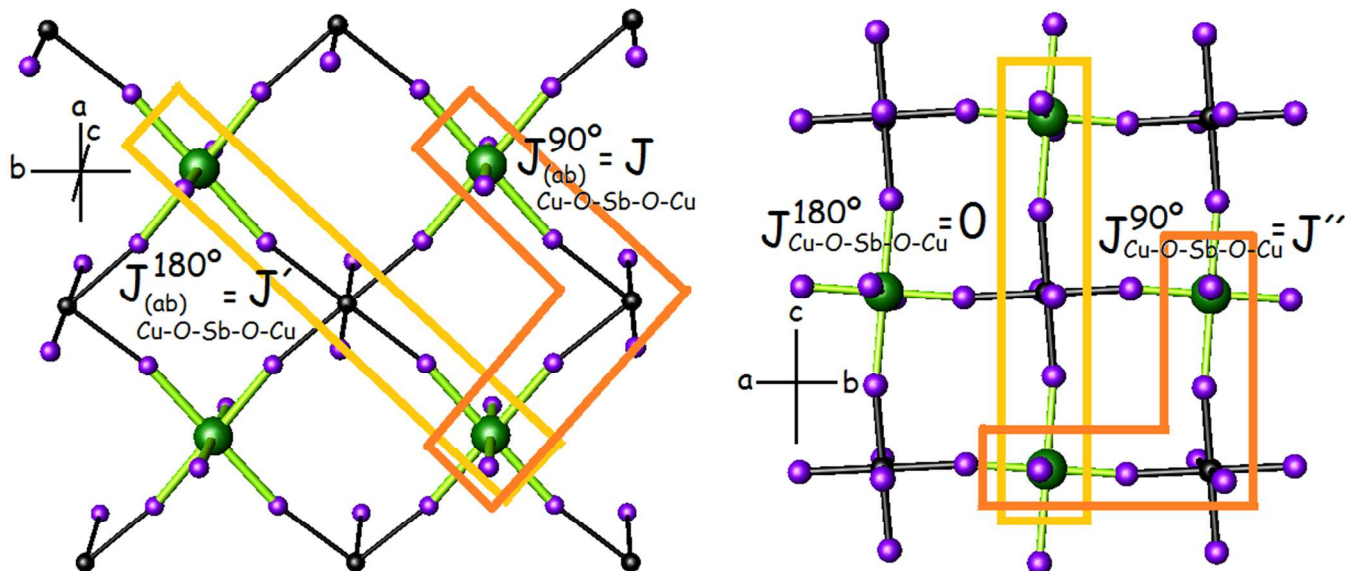
[9] do not calculate J and J' values for  $\text{Sr}_2\text{Cu}(\text{W}_{1-x}\text{Mo}_x)\text{O}_6$ , but they have discussed the frustrated nature of these systems.

## Discussion

The magnetic orbital, for  $\text{Cu}^{2+}$  ion, is the half filled  $d_{x^2-y^2}$ , and can be considered to lie on the *ab* plane for the perovskite  $\text{BaLaCuSbO}_6$ . On the other hand, the filled  $d_z^2$  orbital is not magnetic and interacts with the oxygen  $p_z$  orbitals along the *c*-axis, and consequently, there is not magnetic interaction along this direction. Because of this, the low dimensional magnetic behaviour can be considered mostly confined to the *ab* planes and the material can be considered to be a two dimensional Heisenberg antiferromagnet of spins  $1/2$ .

The two  $\text{Cu}^{2+}$  ions that lie on the *ab* plane, as illustrated in **Figure 6** (left panel), are separated by 7.87 Å in the  $\text{Cu}^{2+}\text{--O}^{2-}\text{--Sb}^{5+}\text{--O}^{2-}\text{--Cu}^{2+}$  paths at  $180^\circ$  ( $J_{(ab)}^{180^\circ}$  or J'), and 5.57 Å in the  $\text{Cu}^{2+}\text{--O}^{2-}\text{--Sb}^{5+}\text{--O}^{2-}\text{--Cu}^{2+}$  paths at  $90^\circ$  (when they are the nearest neighbor magnetic ions) ( $J_{(ab)}^{90^\circ}$  or J). But, if interplanar interactions are taken into account, see **Figure 6** (right panel), then, the distances are bigger, 8.46 Å and 5.77 Å, respectively. Thus, there are four possible magnetic interactions; two of them, previously defined, called  $J_{(ab)}^{90^\circ}$  and  $J_{(ab)}^{180^\circ}$ , showed in **Figure 6** (left panel) and the two interplanar paths called  $J^{90^\circ}$  and  $J^{180^\circ}$ , **Figure 6** (right panel).

**Figure 6:** Proposed magnetic interactions for BaLaCuSbO<sub>6</sub> DP: In a square lattice of magnetic Cu<sup>2+</sup> ions, left: in *ab* plane, right: along *c* axes.



According to Goodenough-Kanamori rules [27] for magnetic interactions, both the in-plane Cu<sup>2+</sup>-O<sup>2-</sup>-Sb<sup>5+</sup>-O<sup>2-</sup>-Cu<sup>2+</sup> path, corresponding to  $J^{180^\circ}_{(ab)}$ , and Cu<sup>2+</sup>-O<sup>2-</sup>-Sb<sup>5+</sup>-O<sup>2-</sup>-Cu<sup>2+</sup> paths, corresponding to  $J^{90^\circ}_{(ab)}$ , are antiferromagnetic since the magnetic interactions are between orbitals that are each half-filled. The interplanar  $J^{180^\circ}$  interaction is not magnetic and can be ignored. On the other hand, the interplanar  $J^{90^\circ}$  will be dismissed because of the long distance between Cu<sup>2+</sup> ions.

The Cu<sup>2+</sup> ions distribution in *ab* plane can easily be seen like a square lattice, **Figure 6**, left side. In a square lattice like this, the shorter magnetic interaction, between nearest neighbours, is  $J^{90^\circ}$ , which corresponds to  $J$  in **Table 3**. The next nearest neighbour interaction,  $J^{180^\circ}$ , correspond to the larger Cu<sup>2+</sup>-O<sup>2-</sup>-Sb<sup>5+</sup>-O<sup>2-</sup>-Cu<sup>2+</sup> superexchange paths,  $J'$  in **Table 3**. The bigger magnitude reported for  $J^{90^\circ}$  in **Table 3** allows us to characterize the BaLaCuSbO<sub>6</sub> DP as a Neel Antiferromagnet. Thus, the shorter distance for Cu<sup>2+</sup>-O<sup>2-</sup>-Sb<sup>5+</sup>-O<sup>2-</sup>-Cu<sup>2+</sup> at 90° paths compared to the longer superexchange paths Cu<sup>2+</sup>-O<sup>2-</sup>-Sb<sup>5+</sup>-O<sup>2-</sup>-Cu<sup>2+</sup> at 180°, favours  $J^{90^\circ}_{(ab)}$  over  $J^{180^\circ}_{(ab)}$ .

For antiferromagnetic B-site ordered DP Ba<sub>2</sub>CuWO<sub>6</sub>, Todate *et al.* [28] also obtained results that suggest the predominance of superexchange magnetic interaction in the *ab*-plane.

To the best of our knowledge, this is the first time that the different magnetic interactions in a two dimensional antiferromagnetic DP containing Cu<sup>2+</sup> were quantified using an analytical function, obtaining in this way a deeper understanding of the magnetic behaviour of these DP.

## Conclusions

The BaLaCuSbO<sub>6</sub> monoclinic DP, space group I 2/m, has been successfully synthesized by solid-state reaction in air atmosphere. It has 4% *antisite* disorder of B cations and magnetic measurements show 2D-antiferromagnetic behaviour with T<sub>N</sub> around 64 K, followed by a transition from a 2D to a 3D-antiferromagnetic regime. The JT effect of Cu<sup>2+</sup> ions originates the structural features, which favours the in-plane superexchange antiferromagnetic interactions,  $J^{90^\circ}$  ( $J$ ), over the in-plane  $J^{180^\circ}$  ( $J'$ ) antiferromagnetic superexchange interaction. Both,  $J$  and  $J'$  magnetic interactions have been evaluated according to a Heisenberg antiferromagnetic rectangular model using an approximation to Curie's law in powers of  $J/T$ , being  $|J|$  around 10 times stronger than  $|J'|$ . To the best of our knowledge, this is the first time that the different magnetic interactions in a two dimensional antiferromagnetic DP containing Cu<sup>2+</sup> were quantified using an analytical function.

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### Notes and references

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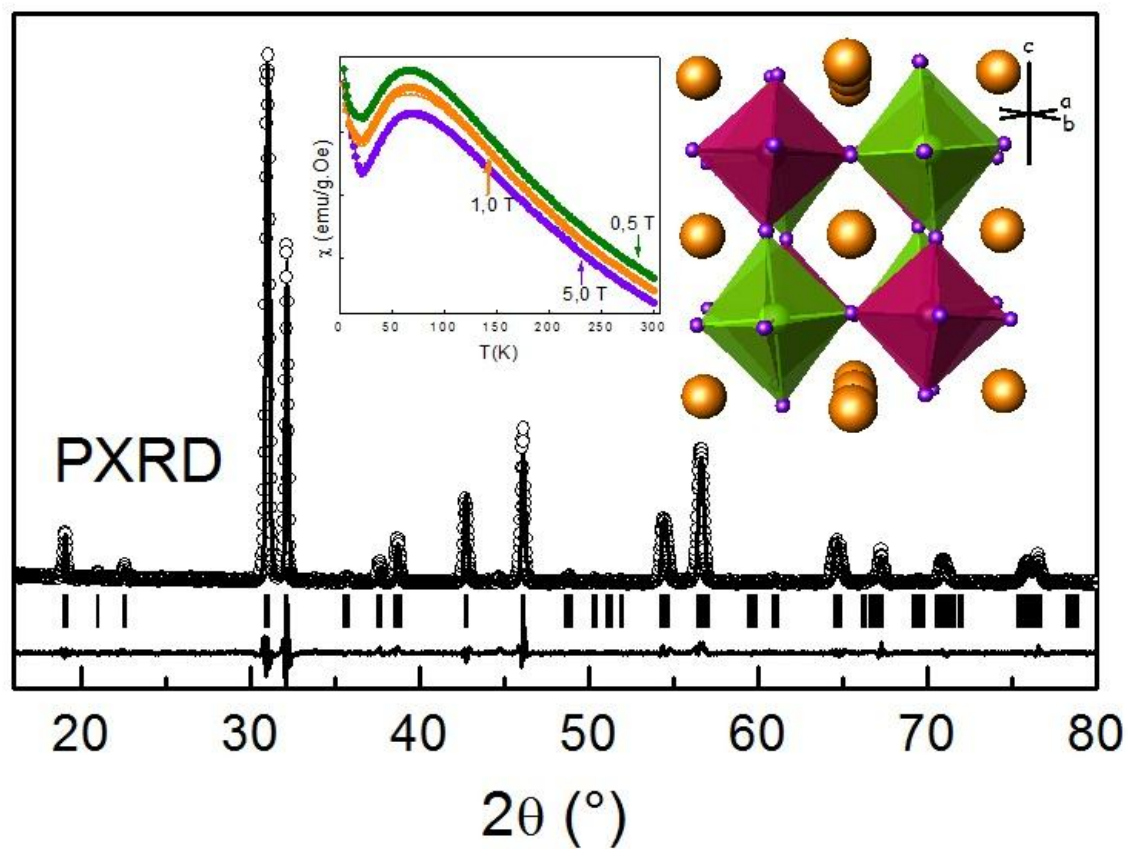
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Electronic Supplementary Information (ESI) available:

Structural information derived from the crystal structure refinement of BaLaCuSbO<sub>6</sub> has been deposited at the ICSD Fachinformationszentrum Karlsruhe (FIZ) (E-mail: [CrySDATA@FIZ.Karlsruhe.DE](mailto:CrySDATA@FIZ.Karlsruhe.DE)), with ICSD file number 428901 (for BaLaCuSbO<sub>6</sub> at 300 K).

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Powder X-ray diffraction pattern, Magnetic Susceptibility as Temperature Function, and polyhedral representation along c-axis of BaLaCuSbO<sub>6</sub> double perovskite.