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## Magnetic susceptibility of one-dimensional spin chains $S(\text{Ni}^{2+}) = 1$ single crystal $\text{Y}_2\text{BaNiO}_5$ in the region of 1.85–800 K

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The magnetic susceptibility of  $\chi(T)$  one-dimensional spin chains (OSCs) of a single crystal  $\text{Y}_2\text{BaNiO}_5$  was first studied in the range of 1.85–800 K. In the region from 800 to  $\sim 520$  the OCC exhibits Curie-Weiss paramagnetism, in which  $\chi_0(T) = C/(T + 800)$ , where  $C$  is the Curie constant, with an effective magnetic moment of  $\mu_{\text{eff}} = 3.75\mu_{\text{B}}$ . With a decrease from 520 to 40 K, the susceptibility changes as a Haldane magnet with a gap  $\Delta = 117$  K,  $\chi(T) = \chi_0(T) \exp(-117/T)$ . Below 40 K  $\chi(T)$  grows again according to the Curie-Weiss law with  $\mu_{\text{eff}} = 3.75\mu_{\text{B}}$ ,  $\theta_{i1} = -3$  K and  $n_{i1} = 9.3 \cdot 10^{19}$  spins  $S(\text{Ni}^{2+}) = 1$  per mole in a crystal grown in an oxygen-free environment; and  $\theta_{i2} = -1.3$  K, and  $n_{i2} = 4.6 \cdot 10^{19}$  spins  $S(\text{Ni}^{2+}) = 1$  after annealing this crystal to 1000°C in air and its subsequent slow cooling. Such a change in the "impurity" contribution to  $\chi(T)$  of the OCC is presumably due to a lack of  $\text{O}_2$  in a significant proportion of the terminal spins in the OCC of a single crystal

**Keywords:** energy gap, SQUID measurements in the region of 1.85–800 K, end spins, breaks in the spin chain.

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### 1. Introduction

The main purpose of the study is to investigate experimentally various types of magnetism occurring with temperature variation in one-dimensional spin chains (OSC) of  $\text{Y}_2\text{BaNiO}_5$  single-crystal, including Curie-Weiss paramagnetism at  $T > 520$  K, Haldane gap magnetism and, finally, low-temperature „impurity“ paramagnetism.

The interest in one-dimensional spin chains has been induced by the well-known study [1] that has promoted the investigations of low-dimensional structures. In 1983 an assumption was put forward [2] that the ground state of integer spin OSC is singlet, separated from the lower excited state by the exchange-origin energy gap  $\Delta$ . As a result, a new theoretical study area has emerged that is focused on the influence of quantum effects in crystal OSC on the macroscopic properties of crystals [3–11]. This, in turn, has promoted experimental study of physical properties [12–30] of such crystals, including: the influence of temperature and gap  $\Delta$  on magnetic susceptibility [15–20,24–28] and inelastic neutron scattering [18–21], magnetic field impact on the gap [14,15], and on other properties, including optical [24,28], thermal [30], electron paramagnetic resonance (EPR) [22,23,29], nuclear magnetic resonance [26].

The first proofs of gap existence  $\Delta$  in OSC were obtained by numeric methods [6,7] and experiments on inelastic neutron scattering on ions  $S(\text{Ni}^{2+}) = 1$  in  $\text{CsNiCl}_3$  [12] and  $\text{Ni}(\text{C}_2\text{H}_8\text{N}_2)_2\text{NO}_2\text{ClO}_4$  [13–15]. However, the major focus was made on the study of OSC in  $R_2\text{BaNiO}_5$  [18–30], where  $R$  is one of rare-earth metal ions or  $\text{Y}^{3+}$ . Special benefit of

such compounds is in that negligible substitutions in them of ions of some metals with ions of other metal considerably modified the crystal properties while maintaining in most cases the one-dimensional characteristic of  $\text{Ni}^{2+}$  spin chains. That is why many researchers (see, for example, [21–23,29]) considered  $\text{Y}_2\text{BaNiO}_5$  as an ideal Haldane magnet. Structure of this crystal corresponds to rhombic crystal system where flattened octahedra  $\text{NiO}_6$  are bound with each other via an oxygen atom and form  $S(\text{Ni}^{2+}) = 1$  one-dimensional spin chains along the crystallographic axis  $\mathbf{a}$ . Unidimensionality of spin chains (OSC) is supported by the fact that intrachain exchange interaction  $J^0$  is more than  $10^3$  times higher than interchain exchange interaction  $J^1$  [10,18].

The temperature dependence of OSC magnetic susceptibility in  $\text{Y}_2\text{BaNiO}_5$  was first studied on a powder sample [18]. Though dependence  $\chi(T)$  was obtained in a wide temperature range (1.85–800 K), the authors of [18] did not address magnetism at  $T > 500$  K, supposing apparently that there was an uncontrolled impurity contribution in it. All following investigations of susceptibility  $\chi(T)$  [20,24–26] were carried out on compact crystals using primarily the temperature range of 300 K and slightly higher. As a result, it was found that, independently of the method of production of  $\text{Y}_2\text{BaNiO}_5$  crystals and the presence of various impurities in them, and on the type of test samples (whether polycrystalline or single-crystal), all samples exhibited the same behavior of dependences  $\chi(T)$ . In particular, at  $T < 300$  K, gradual drop of  $\chi(T)$  occurred, and after passing through the minimum near 40 K, dramatic

growth of  $\chi(T)$  took place almost by the Curie law. Such behavior of  $\chi(T)$  contradicted with the Haldane model [2,3]. This was unsuccessfully explained by the presence of a small fraction of paramagnetic or ferrimagnetic impurity in  $\text{Y}_2\text{BaNiO}_5$  crystals or of  $\text{Ni}^{3+}$  impurity with excess oxygen in the crystal [18,20,24–26].

According to the current theoretical concepts [2–11] and  $\text{Y}_2\text{BaNiO}_5$  crystal structure data [16] in high temperature region, magnetic susceptibility of spins in chains acts as a paramagnetic assembly of weakly interacting  $S(\text{Ni}^{2+}) = 1$  spins obeying the Curie–Weiss law [31]:

$$\chi_0(T) = N_A(\mu_{\text{eff}})^2 \{3k_B(T + \theta_0)\}^{-1}, \quad (1)$$

where  $\theta_0$  is the Weiss temperature,  $N_A$  is Avogadro's number,  $\mu_{\text{eff}} = p_{\text{eff}}\mu_B$  is the effective magnetic moment of  $\text{Ni}^{2+}$ ,  $k_B$  is the Boltzmann constant,  $\mu_B$  is the Bohr magneton.

When  $T$  is lower than some  $T^*$ , arbitrary pairs of adjacent spins in OSC start exhibiting antiferromagnetic interaction in a spontaneous manner and form nonmagnetic  $S(\text{Ni}^{2+}) = 1$  clusters from two spins. These interaction are of indirect nature, because they occur due to  $O^{2-}$   $2p$ -orbitals overlapping with  $\text{Ni}^{2+}$   $3d$ -orbitals [8]. Such spin pairs near  $T^* > T > \Delta/k_B$  form instable singlet states similar to superconducting electron pairs occurring in a spontaneous manner in the normal electronic excitation spectrum above the superconducting gap.

The fraction of spins involved in the OSC magnetism naturally decreases proportionally to the number of spins that retained their paramagnetic state:  $n = N_A \exp(-\Delta/T)$ , and  $\chi(T)$  as written as

$$\chi(T) = N_A(p_{\text{eff}}\mu_B)^2 \exp(-\Delta/T) \{3k_B(T + \theta_0)\}^{-1}. \quad (2)$$

Using equations (2) and (1), it can be shown that

$$\chi(T)/\chi_0(T) = \exp(-\Delta/T). \quad (3)$$

Thus, the experimental susceptibility measurements with temperature decrease below  $T^*$  may be directly used to determine the gap size  $\Delta$  without involving a computer-aided fit technique.

## 2. Experiment

For the purpose of the study, a single-crystal was grown on a nucleus attached to a platinum wire with slow pulling from melt. The melt was prepared from a mixture of  $\text{Y}_2\text{O}_3$ ,  $\text{BaO}$  and  $\text{NiO}$  powders with molar ratio  $\text{Y}:\text{Ba}:\text{Ni} = 5:53.5:41.5$ . A platinum crucible with the powder mixture was placed in a batch furnace that was filled with argon after evacuation. Then, the mixture was heated to  $1100^\circ\text{C}$  to perform a solid-state reaction in them and was held at this temperature during 24 h. For homogenization of the melt, the resulting compound was held during 24 h at  $T = 1450^\circ\text{C}$ . Then, the melt temperature was reduced from  $1450$  to  $1400^\circ\text{C}$  at a rate of  $10^\circ\text{C/h}$  and

then to  $1160^\circ\text{C}$  at a rate of  $1^\circ\text{C/h}$ . The last stage — cooling down to room temperature — was performed at a rate of  $50^\circ\text{C/h}$ . Then, the crystal was carefully washed with a weak acetic acid solution in methanol. A semitransparent crystal had a darkish shade. Structural analysis of the synthesized single-crystal has shown that it has an orthorhombic cell structure with parameters (in Å):  $a = 3.761$ ,  $b = 5.759$  and  $c = 11.330$ , that almost coincide with those obtained earlier in [16]. The crystal volume was  $5 \times 4 \times 1.5 \text{ mm}^3$  and its shape was similar to an elongated prism resembling a hexagon „pencil“. The crystallographic axis of a crystal corresponding to the direction of one-dimensional  $\text{Ni}-\text{O}-\text{Ni}-\text{O}$  chains coincided with the principal axis of the „pencil“. Disorientation of the single-crystal blocks did not exceed  $\pm 0.02^\circ$ . Magnetic measurements were carried out using „Quantum Design“ (MPMS 7.0) SQUID magnetometer in susceptibility mode in the temperature ranges from 1.85 to 400 K (the first range) and 300–800 K (the second interval). These two ranges were „sewn together“ near 300–400 K. single-crystal orientation with respect to the magnetic field was selected such that its crystallographic axis  $\mathbf{a}$  coincided with the magnetic field direction. This allowed to get rid of the impact of  $g$ -factor and demagnetizing factor anisotropy on the magnetic susceptibility of a crystal.

## 3. Finding and discussion

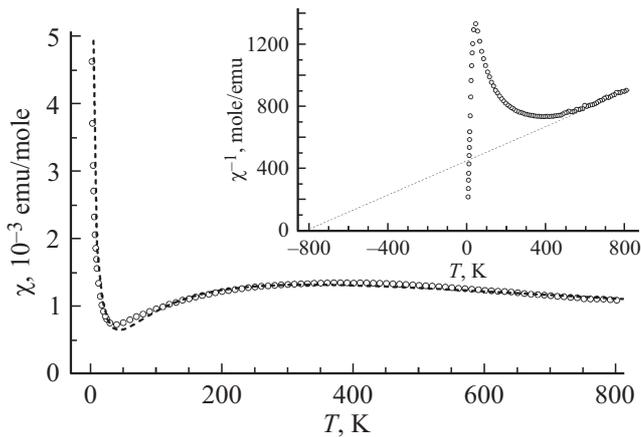
The shape of magnetization curves  $\sigma(H)$  of  $\text{Y}_2\text{BaNiO}_5$  crystal at 1.85 and 800 K in the field up to 5 T corresponded to the Langevin function. This suggested that the magnetization of the single-crystal in question was attributable to its paramagnetic component. A linear segment present when field  $H$  changed from 0 to 0.5 T was specific to dependences  $\sigma(H)$  at any temperature in the range of 1.85–800 K.

Experimental dependence  $\chi(T)$  measured along the crystallographic axis  $\mathbf{a}$   $\text{Y}_2\text{BaNiO}_5$  in magnetic field  $H = 1 \text{ kG}$ , is shown in Figure 1. As mentioned above, similar dependence  $\chi(T)$  was earlier obtained in the same temperature range for a powder sample [18]. It was found that  $\chi(T)$  of the powder sample almost completely coincided with dependence  $\chi(T)$  shown in Figure 1 both in its shape and absolute susceptibility values at the same temperatures. Therefore, it is obvious that paramagnetic susceptibility of OSC in  $\text{Y}_2\text{BaNiO}_5$  crystal is of isotropic type.

The detail in Figure 1 shows that experimental dependence  $\chi(T)$  in coordinates  $\chi^{-1}$  on  $T$  in the range of 800–520 K is almost linear. This is indicative of paramagnetic state of all spins of  $S(\text{Ni}^{2+})$  single-crystal in this temperature range where magnetic susceptibility  $\chi_0$  may be described by the Curie–Weiss law [31], i.e. as

$$\chi_0^{-1} = 3k_B(T + \theta_0)/(N_A\mu_{\text{eff}}^2). \quad (4)$$

From comparison of equation (4) with experimental dependence  $\chi^{-1}$  on  $T$  shown with a dotted line in Detail in Figure 1, it follows that its intersection with the temperature



**Figure 1.** Experimental dependence of magnetic susceptibility  $\chi$  on the temperature measured along the axis **a** of  $\text{Y}_2\text{BaNiO}_5$  crystal. The dotted line shows theoretical dependence  $\chi(T)$  built using equation (8). Detail: dependence of  $\chi^{-1}$  on  $T$ .

axis gives:  $\theta_0 = 800 \pm 50$  K, and magnetic moment of  $\text{Ni}^{2+}$  is equal to  $\mu_{\text{eff}} = (3.75 \pm 0.10)\mu_B$ .

It should be noted that, first, drop of  $\chi(T)$  at  $T < 520$  K may be associated only with initial manifestation of antiferromagnetism in the spin chains of  $\text{Y}_2\text{BaNiO}_5$  crystal, therefore,  $T^* \approx 520$  K may be assumed. Second, a surprising, but indisputable fact is that temperature point  $T^*$  almost coincides with the Neel temperature of  $\text{NiO}$  which has  $T_N = 520$  K [33,34]. This antiferromagnetic interaction in  $\text{NiO}$  is naturally three-dimensional and of cooperative type, while with  $\text{Y}_2\text{BaNiO}_5$  OSC crystal, it exhibits pronounced unidimensionality along the crystallographic axis **a**. Though  $T^*$  is not of cooperative nature, antiferromagnetic (AFM) constants  $J$  of both crystals are have close value:  $|J|/k_B \approx 520$  K.  $T_N$  and  $T^*$  are also common in that in both structures AFM interaction between spins is performed indirectly via  $\text{O}^{2-}$ .

Another feature of the OSC spins is that the experimentally found  $p_{\text{eff}} = 3.75$  obviously exceeded the calculated  $p_{\text{eff}} = 2[S(S+1)]^{0.5} = 2.83$  with full „freezing“ of orbital moment  $L$ . Using the known Stoner relation [32],

$$p_{\text{eff}} = \{[2S(S+1) + \beta L(L+1)]g_{\text{exp}}\}^{1/2}, \quad (5)$$

and experimental values  $p_{\text{eff}} = 3.75$ ,  $g_{\text{exp}} = 2.25$  found by the EPR method along the axis **a** of the crystal [22,28,29], and taking into account the fundamental term of Ni atom corresponds to  ${}^3F_4$ , it can be shown that  $\beta = 0.2$ . This value  $\beta$  shows the degrees of spin-orbital interaction „defrosting“ and its impact on magnetic moment  $\text{Ni}^{2+}$ . It should be noted that  $\text{Ni}^{2+}$  in  $\text{NiO}$  has a similar value  $p_{\text{eff}}$ , and  $\theta$  of this oxide is also higher and is equal to  $-2470$  K.

To find the size of Haldane gap  $\Delta$  (in temperature dimensionality), equation (3) was used which is written as

$$T \ln\{\chi(T)/\chi_0(T)\} = -\Delta. \quad (6)$$

Using the plotted dependence (6) taking into account the scatter of the experimental points, rms error was assessed:  $\Delta_0 = 117 \pm 3$  K.

Compare experimentally the found value  $\Delta_0$  with the gap found earlier in [18], where  $\text{Y}_2\text{BaNiO}_5$  powder sample was used. The gap was assessed as  $\Delta_1 = 97$  K. It was also calculated from the experimental magnetic susceptibility data  $\chi(T)$ , but by computer-aided fitting of a three-term analytical expression containing  $\Delta$  and two other variables to the experimental dependence  $\chi(T)$ .

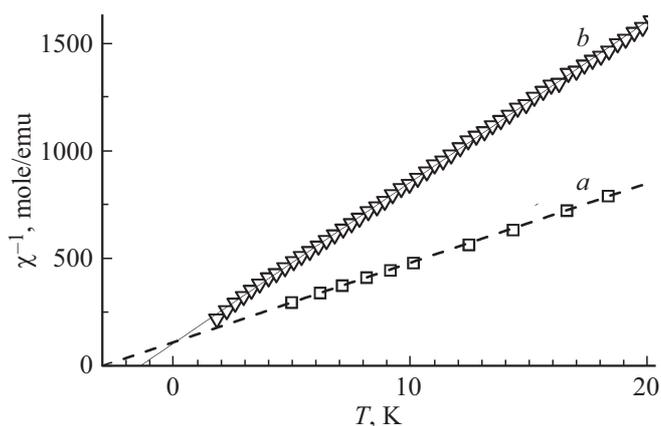
Also, [18] provides inelastic neutron scattering data obtained on the same  $\text{Y}_2\text{BaNiO}_5$  powder sample. In the neutron scattering spectrum, washed-out neutron scattering intensity peaks were detected. The authors suggested that the most intense peak is associated with the Haldane gap size and assumed  $\Delta = 127 \pm 10$  K.

Later, Japanese researchers have carried out similar investigations of the gap size in the energy spectrum of one-dimensional spin chains in  $\text{Y}_2\text{BaNiO}_5$  composite crystal bonded of two single-crystals [20]. they also used the computer-aided fitting method similar to that used in [18] to find gap size  $\Delta$ . The gap determined using the magnetic susceptibility along the crystallographic axis of **a** crystal corresponded to  $\Delta_{\parallel} = 118$  K, and the perpendicular to this axis was  $\Delta_{\perp} = 103.9$  K. The investigations of inelastic neutron scattering have shown the averaged  $\Delta_n = 108$  K. In conclusion of [20], the authors state that the spin gap in  $\text{Y}_2\text{BaNiO}_5$  crystal is isotropic and is approximately equal to  $\Delta = 110$  K.

It has been mentioned above that dependences  $\chi(T)$  of any  $\text{Y}_2\text{BaNiO}_5$  samples at  $T < 40$  K show intense growth of  $\chi(T)$  by the Curie law [18–20,24–28]. This has been generally explained by the introduced paramagnetic and ferrimagnetic impurity. But the phenomenon of abnormal growth of  $\chi(T)$  below 40 K may be associated with the nature of OSC, more specifically, with the spin chain length limitation. This phenomenon has been first addressed in [18]. Independently of the number of spins in OSC, adjacent spin pairs are capable of forming stable nonmagnetic singlets at  $T < \Delta$ . Therefore, only OSC end spins containing  $S(\text{Ni}^{2+}) = 1$  spins in their ends can contribute to the magnetism of  $\text{Y}_2\text{BaNiO}_5$  crystal.

Microscopic investigations of  $\text{Y}_2\text{BaNiO}_5$  crystal grown in inert medium with surface washed with methanol/acetic acid solution have detected many microcracks on the surface. It has been decided to check the influence of crystal annealing in air on the low-temperature magnetic susceptibility of the crystal.

Linear dependences  $\chi^{-1}(T)$ , when  $T < 20$  K shown in Figure 2 show that susceptibility growth in this temperature region is associated with a paramagnetic impurity whose nature seem unclear at first sight. However, comparison of low-temperature segments of dependences  $\chi^{-1}$  on  $T$  for the single-crystal immediately after completion of its growth (Figure 2, a) and after annealing in air (Figure 2, b) suggested that the paramagnetic impurity concentration depends on the single-crystal defects induces by the stresses



**Figure 2.** low-temperature dependences  $\chi^{-1}(T)$  at  $T < 20$  K for  $\text{Y}_2\text{BaNiO}_5$  crystal: *a* — grown in argon atmosphere and *b* — after annealing in air at  $1000^\circ\text{C}$ .

in OSC. Linearity of the dependences indicates that they vary according to the Curie–Weiss law in this temperature range.

$$\chi^{-1} = 3k_{\text{B}}(T + \theta_{i1}) / (n_{i1}\mu_{\text{eff}}^2) \quad (7)$$

with the following parameter values: for air-annealed crystal  $\theta_{i2} = -1.2$  K;  $n_{i2} = 4.6 \cdot 10^{21}$ , and for initial crystal  $\theta_{i1} = -3.0$  K;  $n_{i1} = 9.3 \cdot 10^{21}$ .

Low-temperature value  $\mu_{\text{eff}}$  shall not apparently differ greatly from its high-temperature value. Therefore,  $n_i$  and  $\theta_i$  nominally assigned to „impurity“ paramagnetism of  $\text{Y}_2\text{BaNiO}_5$  single-crystal calculated using expression (7) show that the concentration of paramagnetic „impurity“ in the initial crystal is approximately twice as high as in an air-annealed crystal, and  $\theta_i$  values are almost twice as low. Fraction of low-temperature paramagnetic „impurity“ particles in the initial single-crystal was 1/60 of the total number of nickel ions and their concentration in single-crystal after annealing in oxygen was almost twice as low and approached 1/120.

Possible origin of „impurity“ paramagnetism will be mentioned briefly below. In OSC at  $T < T^*$ , AFM clusters may appear spontaneously. In this case, mechanical stresses and even chain discontinuities with partial loss of  $\text{O}_2$  may certainly occur in the spin chain.

One of the assumptions on the occurrence of OSC discontinuities in  $\text{Y}_2\text{BaNiO}_5$  crystal is apparently associated with structural stresses induced in them like in  $3d$ -metal oxides (FeO, CoO and NiO) during transition to antiferromagnetic state. Cubic  $\text{O}_h^2$ -lattices of this oxides below the Neel temperature  $T_{\text{N}}$  acquire rhombohedral (in FeO and NiO) and tetragonal (in CoO) distortions (see Table 22.1 in [33] and Table 28.1 in [34]).

Therefore, a sufficient number of end ions that are free from a neighbor capable of forming an antiferromagnetic pair occurs, because now they are located at a distance which is significantly larger than the interionic distance of nickel in OSC. After annealing in air a part of discontinuities

in the chains may be „healed“. Therefore, paramagnetic susceptibility of OSC in  $\text{Y}_2\text{BaNiO}_5$  crystal after annealing in air shall decrease. But apparently it will be difficult to do away with end spins in the crystal OSC, because transition of OSC spin pairs in  $\text{Y}_2\text{BaNiO}_5$  crystal to AFM state at  $T \approx \Delta/k_{\text{B}}$  shall be often followed by spin chain discontinuity.

## 4. Conclusion

It has been found that at  $800$ – $520$  K, one-dimensional spin chains in  $\text{Y}_2\text{BaNiO}_5$  single-crystal exhibit paramagnetic properties whose magnetic susceptibility varies according to the Curie–Weiss law:

$$\chi_0(T) = N_{\text{A}}(3.75\mu_{\text{B}})^2 / 3k_{\text{B}}(T + 800)^{-1}.$$

Below  $520$  K and up to  $\Delta/k_{\text{B}}$ , instable pair antiferromagnetic interactions may occur spontaneously between adjacent  $S(\text{Ni}^{2+}) = 1$  spins in the chains. As a result, adjacent spin pairs in each spin chain can form nonmagnetic clusters. Therefore, a fraction of paramagnetic spins starts decreasing according to law

$$\chi(T) = C \exp(-\Delta/k_{\text{B}}T)(T + 800)^{-1},$$

where  $\Delta/k_{\text{B}} = 117$  K is an experimentally found Haldane gap,  $C$  is the Curie constant.

Intense growth of paramagnetic susceptibility of  $\text{Y}_2\text{BaNiO}_5$  single-crystal at  $T < 40$  K is presumably induced by OSC end spins according to the Curie–Weiss law where  $\mu_{\text{eff}} = 3.75\mu_{\text{B}}$ , and  $\theta_{i1} = -3$  K for the initial sample and  $\theta_{i2} = -1.2$  K after annealing of the same single-crystal at  $1000^\circ\text{C}$  in air and slow cooldown at a rate of  $1^\circ\text{C}/\text{min}$ . The number of end spins after annealing decreased by a factor of two from  $n_{i1} = 9.3 \cdot 10^{19} \text{ M}^{-1}$  to  $n_{i2} = 4.6 \cdot 10^{19} \text{ M}^{-1}$ .

Temperature dependence  $\chi(T)$  of  $\text{Y}_2\text{BaNiO}_5$  single-crystal in the range of  $1.85$ – $800$  K after annealing in air may be described by the following analytical expression:

$$\chi(T) = C \{ (1 + \exp(-117/T))(T + 800)^{-1} + n_{i2}/N_{\text{A}}(T + 1.2)^{-1} \}, \quad (8)$$

here  $C = N_{\text{A}}(\mu_{\text{eff}})^2 / 3k_{\text{B}}$ ;  $\mu_{\text{eff}} = 3.75\mu_{\text{B}}$ ,  $n_{i2}$  is the number of end spins.

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## Conflict of interest

The authors declare that they have no conflict of interest.

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