Electronic supplementary information for: "Evidence of a correlation between magnetic and structural transitions in $Y_{2-x}Zn_xRu_2O_7$ pyrochlore compounds"

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Zero field μSR measurements and analysis

To study the effect of the Zn doping on the antiferromagnetic phase, we performed muon spin relaxation (μSR) measurements on the $Y_{1.80}Zn_{0.20}Ru_2O_7$ sample in conditions of zero-magnetic field (ZF). In a typical ZF-μSR experiment a beam of positive muons 100% spin-polarized along the beam direction is implanted in the sample.[1] Once muons thermalize at interstitial sites (almost instantaneously and without loss of polarization), they act as sensitive probes, precessing in the local magnetic field. The observed quantity is the time evolution of the muon-spin polarization $P_z(t)$, obtained by studying the angular distribution of the positrons emitted during the muon decay process (muon lifetime $\tau_\mu \approx 2.2\mu s$). The time dependence of the muon polarization is displayed in Fig. 1 for $Y_{1.80}Zn_{0.20}Ru_2O_7$ at selected temperatures. The data fit well to the following equation:

$$P_z(t) = (1 - V_{AF})\, P_z^{PM}(t) + V_{AF} P_z^{AF}(t) \tag{1}$$

where

$$P_z^{PM}(t) = e^{-\frac{\sigma_{PM}^2 t^2}{2}} \tag{2}$$

$$P_z^{AF}(t) = \sum_{i=1}^{2} w_i \left[\frac{2}{3} \cos\left(2\pi\gamma_\mu B_{\mu,i}\right) e^{-\frac{\sigma_i^2 t^2}{2}} + \frac{1}{3} e^{-\lambda_i t} \right] \quad . \tag{3}$$

This expression accounts for a paramagnetic $P_z^{PM}(t)$ plus an antiferromagnetic $P_z^{AF}(t)$ terms, being V_{AF} the magnetic volume fraction. In the paramagnetic phase the initial full polarization is mainly preserved for $t \leq \tau_\mu$, being affected only by the weak dipolar interaction of the $\frac{1}{2}$ muon spins with the nuclear magnetic moments which is described by a gaussian relaxation with time constant $\sigma_{PM} \sim 0.01\mu s^{-1}$. The $P_z^{PM}(t)$ term (Eq. **??**) describes the muon polarization well above the magnetic transition T_N (where $V_{AF} = 0$), as shown for $T = 100$ K in Fig. 1. In the magnetically ordered phase of a polycrystalline sample, on average, two-thirds (one-third) of the incident muon polarization will lie perpendicular (parallel) to the local field. The two-thirds component in Eq.3 reflects the muon spin precession around the local field at the muon site, B_μ proportional to the staggered magnetization, by a damped oscillation function with angular frequency $\gamma_\mu B_{\mu,i}$ for each inequivalent muon site i (being γ_μ=136 MHz/T the muon giromagnetic ratio). The gaussian depolarization time of this component is a measure of the second moment of the internal field distribution at

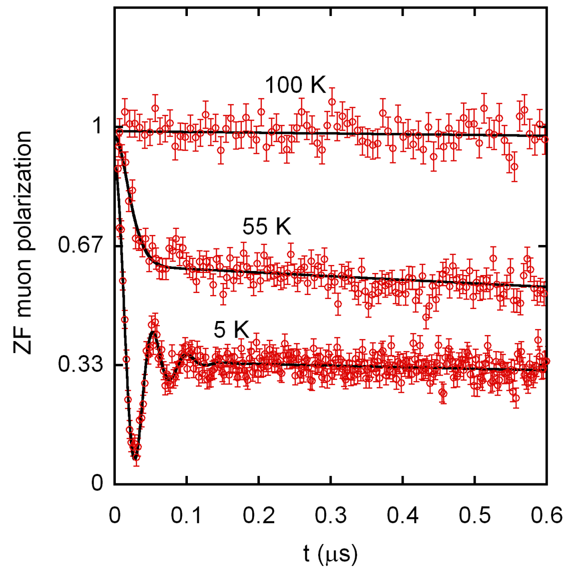

FIG. 1. Typical time dependence of the muon polarization for temperature well below, nearly at and well above the magnetic temperature T_N for $Y_{1.80}Zn_{0.20}Ru_2O_7$. The lines are the best fit to Eq. 1.

the i-site $\Delta B_{\mu,i} \equiv (\overline{B_{\mu,i}}^2)^{1/2} = \sigma_i/\gamma_\mu$. The one-third component in Eq. 3 simply decays exponentially with a decay constant λ_i that accounts for spin-lattice relaxation processes. We found that the $P_z^{AF}(t)$ term describes pretty well ($\chi^2 \cong 1$) the behavior at low temperature, well below T_N, as shown for $T = 5$ K in Fig.1. The weight w_i is the muon fraction occupation of the i-site. Across the AFM-PM transition we expect that the paramagnetic phase gradually appears at expenses of the magnetic volume and the muon polarization function is well fitted by Eq. 1 with $0 < V_{AF} < 1$ (e.g. solid line for $T = 55$ K in Fig.1). In this regime the oscillations get overdamped and disappear in the muon polarization function, i.e. the field distribution width becomes greater than the local field $\Delta B_\mu^i > B_\mu^i$.

In Fig. 2a the temperature dependence of the magnetic volume fraction, $V_{AF}(T)$, shows that the magnetic transition onset is close to $T_{N,onset} \approx 70$ K. By assuming a gaussian distribution of local magnetic transitions we can fit $V_{AF}(T)$ to an erf-like function (solid line in Fig. 2a) which yields to an average $T_{N,av} = 56 \pm 1$ K with a standard deviation $\Delta T_N = 6 \pm 1$ K.

Our data analysis at low temperatures requires two different frequencies in agreement with previous results on the undoped $Y_2Ru_2O_7$ compound,[2] indicating that two magnetically

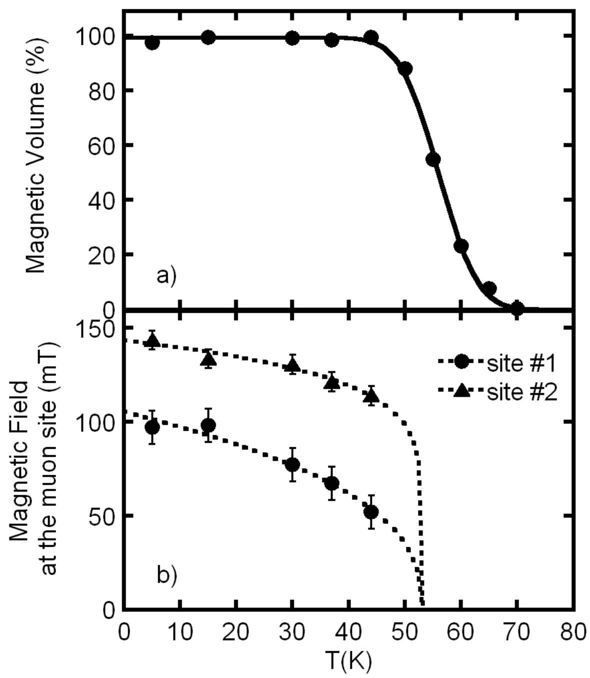

FIG. 2. Temperature evolution of a) the magnetic volume fraction and b) internal field at the muon site. The solid line is a fit to an *erf*-like function and the dotted lines are guides to the eye.

inequivalent muon sites are present. The weights w_i of the two components were fixed over the measured temperature range to be $w_1 = 2.2w_2$. The behavior of the local magnetic fields B^i_μ for $i = 1, 2$ is displayed as a function of temperature in Fig. 2b. The higher local field yields to nearly the same precession frequency found in the undoped ($\gamma_\mu B_{\mu,2} \approx 21$ MHz)[2] while the lower field, with more than double the spectral weight, is reduced by a factor ≈ 1.5. One can notice that also the mean value of the magnetic transition is reduced by almost the same factor. As already mentioned the oscillations get overdamped when approaching the magnetic transition, actually just above the temperature of 45 K at which the magnetic volume fraction starts to decrease, and the internal muon field cannot be further measured. This behavior is markedly different from the one detected in the undoped compound where muon precession frequencies could be finely measured until they approached zero (see Fig. 4 of Ref. 2). The latter behavior is expected when the magnetic transition is quite sharp while the former one is indicative that the Zn doping broadens the transition width.

Summarizing, the effect of the Zn doping as studied by ZF-μSR in the $Y_{1.80}Zn_{0.20}Ru_2O_7$

is to weaken the magnetic ordering, by lowering both the $T_{N,av}$ and the internal local field of the most populated muon site and also by broadening the transition width.

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