between the Mo and Nb nuclei. However this is small compared to the experimental error in the sample mass which is known with 10% accuracy. For this reason the heat capacity was measured well into the paramagnetic regime and matched to the heat capacity of Ba_2YNbO_6 above 200 K (150 K) for the zero-field (9 T) measurements [30]. The magnetic entropy is gradually released over a wide range of temperatures with a broad maximum around 50 K. No anomalies corresponding to phase transitions are observed, only a gradual freezing, quenching all degrees of freedom associated with the orbitally degenerate t_{2g} $s = 1/2$ $4d$ electrons. As shown in the inset of Fig. 4, the total entropy recovered $S_{\rm tot} = 12 \pm 2$ JK^{-1}mol^{-1}, close to the $R \ln 4 = 11.5$ expected for a $j = 3/2$ quadruplet (the $j = l - s = 1/2$ doublet lies at much higher energies [25, 26]). Below 25 K only $\sim 5\%$ of the entropy is released, in agreement with the Curie fit to the low temperature susceptibility which was found to correspond to $\sim 5\%$ of the Mo^{5+} if these remaining spins have $j = 3/2$. In a 9 T magnetic field most of the magnetic entropy shifts to lower temperatures.

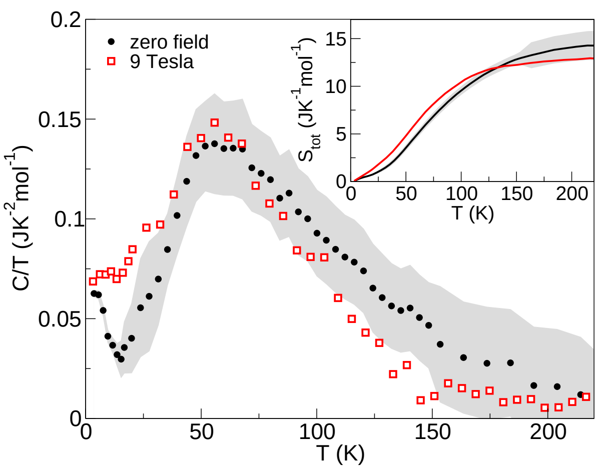

FIG. 4: (Color online) The magnetic heat capacity obtained by subtracting the heat capacity of the diamagnetic analogue Ba_2YNbO_2 in zero field (black dots) and in 9 Tesla (open squares). The experimental error for the 9 T data is comparable to that indicated for the zero field data (grey area). The inset shows the total entropy release as a function of temperature in zero field (black line) and in 9 T (red line).

To gain a better understanding of the gradual freezing and the appearance of apparently weakly-coupled spins a μSR experiment was carried out. The zero-field muon spin relaxation spectra at 120 K, 5 K and 1.4 K are shown in Fig. 5. There is no evidence of muon relaxation due to nuclear spins which confirms that the main muon stopping site is near the O^{2-} ions. At 120 K there is no muon relaxation, as expected for a paramagnetic state. Remarkably, at 5 K a muon relaxation is still only just detectable. If the maximum in the AC susceptibility is due to a conventional spin-glass transition

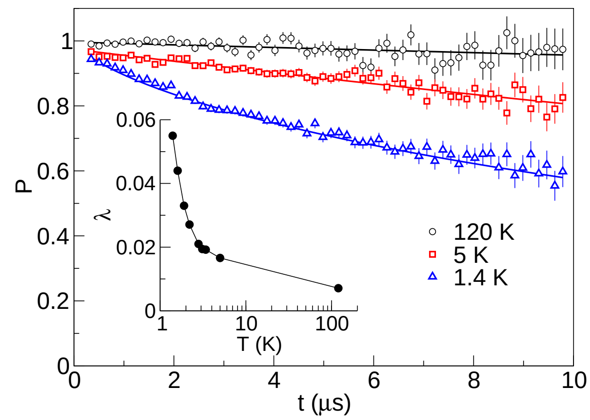

FIG. 5: (Color online) The muon spin relaxation at 120, 5 and 1.4 K. The relaxation follows an exponential decay ($P(t) = \exp[-t/\lambda]^{\beta}$, solid lines) with $\beta = 1$ for all but the 1.4 K data, where $\beta = 0.7$. The temperature dependence of the relaxation rate λ is shown in the inset.

a Lorentzian Kubo-Toyabe muon relaxation is expected below the spin glass transition, as observed in the related system Sr_2MgReO_6 [23]. The very slow muon relaxation observed at 5 K in Ba_2YMoO_6 indicates there are no static moments. At the same time the heat capacity data shows that at 5 K most of the magnetic entropy associated with $j = 3/2$ is quenched, implying static order. The majority of spins must therefore have bound into (non-magnetic) static spin-singlet "valence bonds" in which also the orbital degrees of freedom are quenched. The moderate increase in the muon relaxation rate below 5 K is then due to slowing-down of a small fraction of the spins which are left isolated as domain walls or defects in a (disordered) valence bond crystal (VBC). The best characterization of this state is probably a valence bond glass (VBG) as described in Ref. 17.

The magnetic properties of Ba_2YMoO_6 are very different to those of the related compound Sr_2MgReO_6 [23], where a first order transition to a conventional spin glass state is observed. That the crossover in Ba_2YMoO_6 is not a conventional spin-glass transition is also clear from the unusual frequency dependence of the AC susceptibility. The gradual freezing and cross-over region around 50 K are consistent with a pseudogap predicted for the VBG [17]. This gap, which corresponds to a spin-singlet dimerization energy scale, is filled by levels corresponding to emergent weakly-coupled spins which give rise to a diverging susceptibility as the temperature is decreased. In close agreement with Ref. 17 the observed low temperature susceptibility follows a power law $\chi \propto (T - T_s)^{-\gamma}$ with $\gamma \approx 1$. As noted earlier, this contribution from effectively weakly-coupled spins can not be related one-to-one to any structural disorder but arises as a cooperative effect, due to the amorphous arrangement of spin-singlets. The heat capacity does not become zero at the lowest temperature measured which is consistent with a small