NEXT_NAMASSTE

NEXT_NanoMagnets for quantum SEnsing and Data SToragE

RL: F. Brero
RN: A. Lascialfari

General information

Project duration:
2025-2027

INFN Units:
BO, FI, MI, PV

External collaborators :
Manuel Mariani, INFN-MI and UNIPV
Paolo Santini, UNIPR and INFN-PR

FTE Pavia 2025		1.2
Brero Francesca **RL**	RTD-A UNIPV	0.3
Filibian Marta	Tec. UNIPV	0.2
Lascialfari Alessandro **RN**	PO UNIPV	0.4
Robustelli Test Agnese	PhD UNIPV	0.3
Giroletti Elio	??	??

Results from NAMASSTE (same Units, RN M. Mariani)

Quantum sensing by NMR and SQUID

Magnetic techniques for quantum sensing using single molecule magnets

(PRL, to be submitted)

Single crystals of Mn12-tbu
A.-L- Barra et al., JACS 129, 10754 (2007)

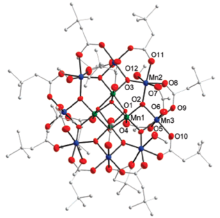
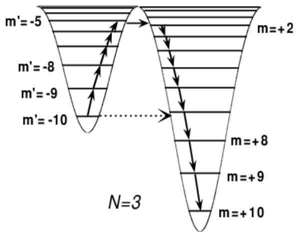


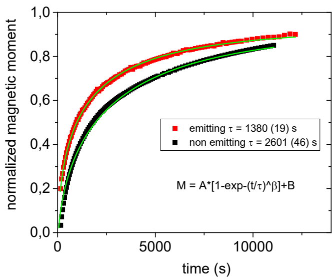
Figure 1. ORTEP view of the molecular structure of $[Mn_{12}O_{12}(Bu{-}CH_2{-}CO_2)_{16}(CH_3OH)_4]\cdot 2CH_3OH$. Mn^{III} sites are reported in blue, Mn^{IV} in green, oxygen in red, and carbon atoms in pale gray. Three *tert*-butyl groups in the labeled region have been omitted for clarity sake.

$$[Mn_{12}O_{12}(O_2CCH_2Bu^t)_{16}(CH_3OH)_4]\cdot CH_3OH$$

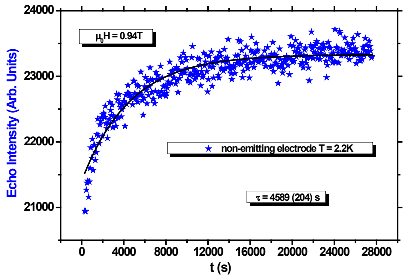


$$\mathcal{H} = -DS_z^2 - BS_z^4 - g_z\mu_B\mu_0 S_z H_z + \mathcal{H}_{trans}$$

Thermal activation :
$\tau_{Magn} = \tau_0 \exp[-\Delta/(k_B T)]$
Quantum tunneling : τ_T

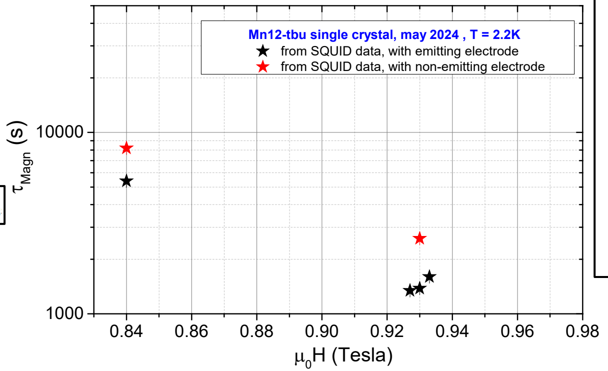


M recovery :
SQUID
NMR

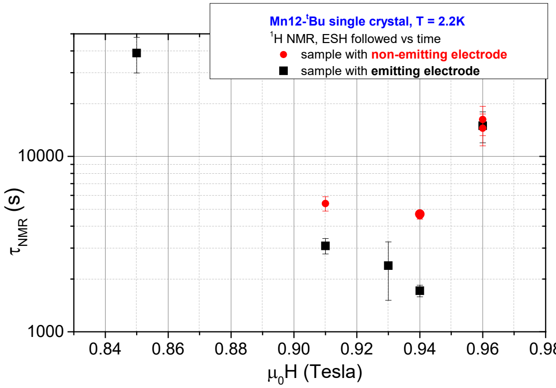


In SQUID and NMR experiments, at B_0~0.94(1) T **we observed a difference in the magnetization recovery time among the cases of ionizing particles impinging and non impinging**.

Recovery time τ vs field, by SQUID



Recovery time τ vs field, by NMR



Results from NAMASSTE (same Units, RN M. Mariani)

Spin dynamics by NMR and DC magnetization

On the spin dynamics of Ln-based single ion magnets : magnetization slowing down and low temperature energy gap

(PRB, to be submitted)

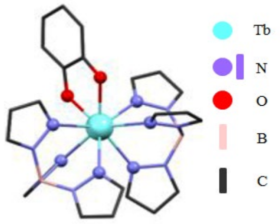


Fig. 1 Structure of the Ln(DTBSQ)(HBPz3)2, Ln-SQ system, where Ln=Tb,Dy

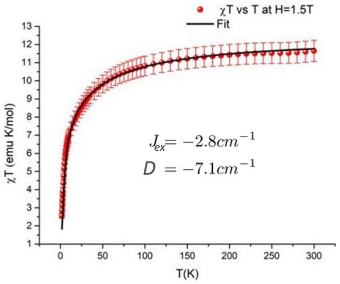
	$\lvert J_{z,Tb}\rvert$	Spin orientation	Energy (cm^{-1})	Energy (K)
E0	6	antiparallel	0	0
E1	6	parallel	36	52
E2	5	antiparallel	231	332
E3	5	parallel	267	384
E4	4	antiparallel	420	604
E5	4	parallel	456	656
E6	3	antiparallel	567	816
E7	3	parallel	603	868
E8	2	antiparallel	672	967
E9	2	parallel	708	1019
E10	1	antiparallel	735	1058
E11	0	antiparallel	756	1088
E12	1	parallel	771	1109
E13	0	parallel	792	1140

Energy levels of Tb-SQ

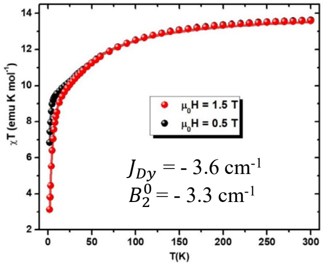
	$\lvert J_{z,Tb}\rvert$	Spin orientation	Energy (cm^{-1})	Energy (K)
E0	15/2	antiparallel	0	0
E1	15/2	parallel	54.2	78
E2	13/2	antiparallel	142.3	204.7
E3	13/2	parallel	191.5	275.6
E4	11/2	antiparallel	264.3	380.3
E5	11/2	parallel	309	444.6
E6	9/2	antiparallel	366	526.5
E7	9/2	parallel	406.7	585.3
E8	7/2	antiparallel	447.	643.1
E9	7/2	parallel	484.9	697.7
E10	5/2	antiparallel	506.7	729.1
E11	5/2	parallel	543.8	782.4
E12	3/2	antiparallel	544	782.8
E13	1/2	antiparallel	556.9	801.2
E14	3/2	parallel	583.9	840.1
E15	1/2	parallel	606.6	872.7
E16	1/2	antiparallel	613.7	883

Energy levels of Dy-SQ

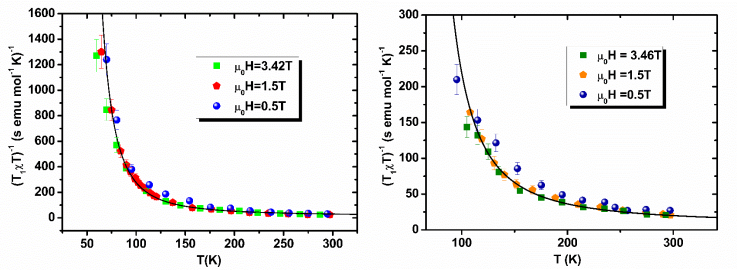
Tb-SQ



Dy-SQ



Tb-SQ Dy-SQ



(a) (b)

Fig. 4 Spin lattice relaxation rate $1/T_1$ data divided by (experimental) χT drawn as a function of temperature at different applied fields, for TbSQ (a) and DySQ (b). The solid lines are fit to the data, obtained following the model cited in the text.

^{1}H NMR spin-lattice relaxation "feels" different gaps in Tb-SQ and Dy-SQ (i.e. different correlation times τ_c)

- Tb-SQ : good fit of M vs T. NMR τ_c follows an Arrhenius law with a gap ~ 2nd excited level gap
- Dy-SQ : good fit of M vs T. NMR τ_c follows an Arrhenius law with a gap ~ 4th excited level gap

Results from NAMASSTE (same Units, RN M. Mariani)

Spin dynamics by MUSR and ac χ

μSR evidence of a marked exchange interaction effect on the local spin dynamics of Tb-based single ion magnets

(PRB, to be submitted)

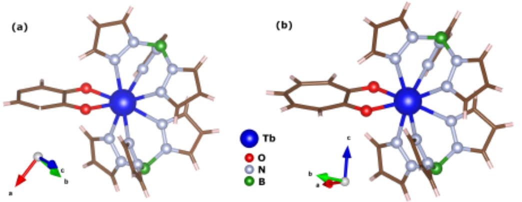
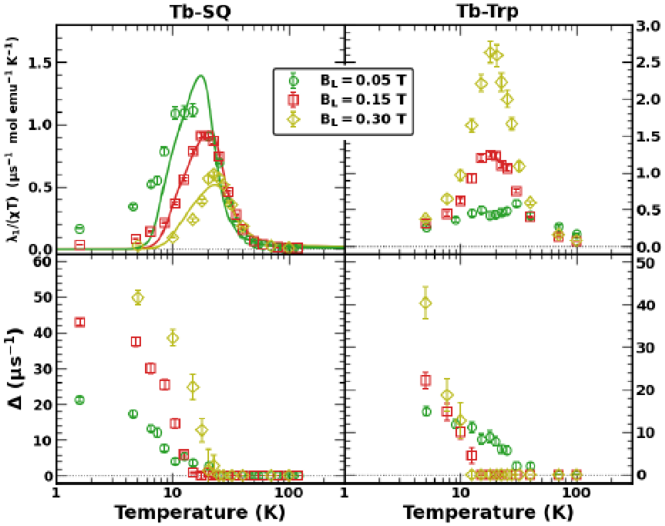


FIG. 1. (Color online) Molecular structure of (a) Tb-SQ and (b) Tt Trp. Hydrogen (pink) and Carbon (brown) are represented in stic mode for the sake of clarity.



$$\lambda_1 \equiv \left(\frac{1}{T_1}\right)_{BPP} = \int_{-\delta}^{+\delta} \rho(E_A)\lambda_1(E_A)dE_A$$
$$= K \cdot \frac{1}{2\omega_L \ln b}\left[\arctan(b\omega_L\tau_c) - \arctan\left(\frac{\omega_L\tau_c}{b}\right)\right],$$

Tb-SQ and Tb-trp show opposite effects on muon Intensity relaxation rates !!

- Tb-SQ : in presence of exchange interaction $\Rightarrow$ BPP law, with distribution of correlation times
- Tb-trp : effect of the “absence” of exchange interaction on spin dynamics (and magnetization slowing down)
- Thus, peculiar field effect on the relaxation time τ for Tb-trp

Planned activities for NEXT_NAMASSTE. I

Quantum Sensing of particles and radiation

- Mn12- tbu sample, B//c : use of proper shielding for α (combined **effect of impinging β particles and γ radiation)** and for β (**effect of γ radiation only**) particles (NMR, SQUID, continuous and pulsed EPR)
- Mn12-tbu sample, **(B_{ac},c) ~10° & (B_{DC} ,c) ~10°** : **effect on MUSR** spectra and **SQUID** magnetization (ν_{ac}=1-1000 Hz)
- Mn12-tbu sample : **MUSR relaxation at T<1K** ($B=B_{cross}$), **with and without impinging particles**
- Development of theoretical models **to simulate the impact of radiation** on Molecular Nanomagnets spin dynamics

$[Mn_{12}O_{12}(O_2CCH_2Bu^t)_{16}(CH_3OH)_4] \cdot CH_3OH$

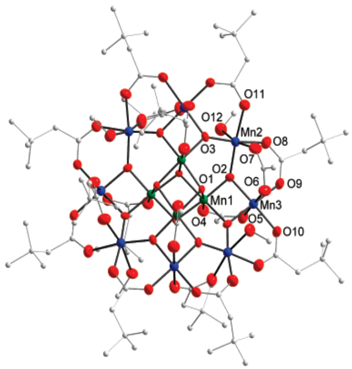


Figure 1. ORTEP view of the molecular structure of $[Mn_{12}O_{12}({}^tBu-CH_2-CO_2)_{16}(CH_3OH)_4]\cdot 2CH_3OH$. Mn^{III} sites are reported in blue, Mn^{IV} in green, oxygen in red, and carbon atoms in pale gray. Three *tert*-butyl groups in the labeled region have been omitted for clarity sake.

Planned activities for NEXT_NAMASSTE. II

Spin dynamics vs temperature (data storage)

- [DyNITpPy]2: DyNIT **units coupled by small (bias) interaction** (tunneling in zero field reduced, slow relaxation favoured), MUSR+NMR
- [Dy(18-C-6)(1-AdO)2][I3] : a system based on a **single Ln ion BUT with a higher barrier**, MUSR+NMR
- **Very low-frequency (ν <5 MHz) spin dynamics by NMR-FFC**, of diluted systems TbSQ, Tb-trp, DySQ, Dy-trp

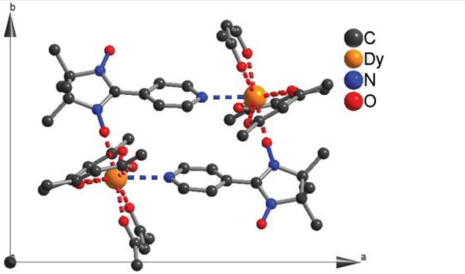
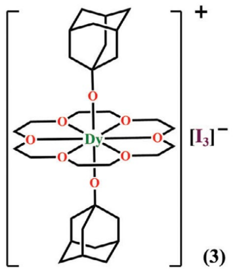


Fig. 1 Schematic view of the $[DyNITpPy]_2$ structure along the crystallographic *c* axis. Fluorine and hydrogen atoms are not shown for the sake of clarity. Dotted lines highlight Dy(III) coordination bonds.

A rational approach to the modulation of the dynamics of the magnetisation in a dysprosium–nitronyl-nitroxide radical complex†

Giordano Poneti,[a] Kevin Bernot,[ab] Lapo Bogani,[a] Andrea Caneschi,[a] Roberta Sessoli,*[a] Wolfgang Wernsdorfer[c] and Dante Gatteschi[a]

Chem. Commun., 2007, 1807–1809 | 1807



(3)

$[Dy(18\text{-}C\text{-}6)(1\text{-}AdO)_2][I_3]$

Bis-Alkoxide Dysprosium(III) Crown Ether Complexes Exhibit Tunable Air Stability and Record Energy Barrier

*Wen-Jie Xu, Qian-Cheng Luo, Zi-Han Li, Yuan-Qi Zhai, and Yan-Zhen Zheng**

Adv. Sci. **2024**, *11*, 2308548

Tentative 3-yrs tasks

Synthesis of SMM for sensing	1-24
Synthesis of rare-earth compounds	1-24
NMR experiments with impinging β and/or γ	3-36
SQUID exp.s with impinging β and/or γ	3-36
CW/pulsed EPR exp.s with impinging β and/or γ	6-36
MUSR exp.s with impinging α, β and/or γ	12-36
MUSR exp.s with ac+DC fields applied, B_{ac} & B^{DC} not parallel to c	12-36
SQUID exp.s with ac+DC fields applied, B_{ac} & B^{DC} not parallel to c	6-36
MUSR+NMR on DyNIT units coupled by small (bias) interaction	12-36
MUSR+NMR+magnetometry on high-barrier [Ln(18-C-6)(1-AdO)2][I3]	12-36
NMR-FFC of diluted systems TbSQ, Tb-trp, DySQ, Dy-trp	3-28

Tentative 3-yrs budget Pavia

Anno	Capitolo	kEuro	Motivazione
2025	Instrumentation Consumables Missions	7 20 5	High precision Gaussmeter Cryogenic gases and He/N2 liquid, spare electronics, emitting electrodes Collaboration meetings, MUSR experiments, outreach
2026	Consumables Missions	20 5	Cryogenic gases and He/N2 liquid, spare electronics, emitting electrodes Collaboration meetings, MUSR experiments, outreach
2027	Consumables Missions	20 5	Cryogenic gases and He/N2 liquid, spare electronics, emitting electrodes Collaboration meetings, MUSR experiments, outreach

Other Units (2025)

Bologna (1 FTE), MUSR	
Samuele Sanna, PA UNIBO , **RL**	0.6
Matteo Casadei, postdoc UNIBO	0.2
Muhammad Maikudi Isah, postdoc UNIBO	0.2
Firenze (1.1 FTE), ac/DC SQUID, EPR, pulsed EPR	
Barbagli Giuseppe, Ric. INFN	0.1
Celardo Giuseppe Luca, PA UNIFI	0.1
Cini Alberto, postdoc UNIFI	0
Fittipaldi Maria, PA UNIFI	0.4
Latino Giuseppe, PA UNIFI, **RL**	0.3
Paoletti Simone, Primo Ric. INFN	0.1
Poneti Giordano, RTD-B UNITUS	0.1
Sorace Lorenzo, PA UNIFI	0
Milano (1.1 FTE), MUSR and low-ν NMR	
Paolo Arosio, PA UNIMI, **RL**	0.6
Francesco Orsini, PA UNIMI	0.4
Ivan Veronese, PA UNIMI	0.1

Tentative 3-yrs budget, other Units

Bologna	Capitolo	kEuro	Motivazione
2025	Instrumentation Missions	3 5	Computer dedicated to MUSR data analysis Collaboration meetings/measurements, MUSR experiments, outreach
2026	Missions	5	Collaboration meetings/measurements, MUSR experiments, outreach
2027	Missions	5	Collaboration meetings/measurements, MUSR experiments, outreach

Firenze	Capitolo	kEuro	Motivazione
2025	Consumables Maintenance Missions	17 1.5 2	Liquid helium, reagents, glasses, solvents, spare electronics Instrumentation maintenance Collaboration meetings/measurements, MUSR experiments, outreach
2026	Consumables Maintenance Missions	17 1.5 2	Liquid helium, reagents, glasses, solvents, spare electronics Instrumentation maintenance Collaboration meetings/measurements, MUSR experiments, outreach
2027	Consumables Maintenance Missions	17 1.5 2	Liquid helium, reagents, glasses, solvents, spare electronics Instrumentation maintenance Collaboration meetings/measurements, MUSR experiments, outreach

Milano	Capitolo	kEuro	Motivazione
2025	Consumables Instrumentation Missions	2.5 2 2	Cooling liquids (Galden) and spare electronics and glasses Gaussmeter + dedicated special oscilloscope for FFC-NMR Collaboration meetings/measurements, MUSR experiments, outreach
2026	Consumables Missions	2.5 2	Cooling liquids (Galden) and spare electronics and glasses Collaboration meetings/measurements, MUSR experiments, outreach
2027	Consumables Missions	2.5 2	Cooling liquids (Galden) and spare electronics and glasses Collaboration meetings/measurements, MUSR experiments, outreach