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Exploring the potential of molecular nanomagnets as quantum sensors of radiation



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Alternative detection strategies are under investigation for the development of innovative devices which exploit the extreme sensitivity of quantum systems. These are considered promising for providing new sensors with frontier performances for applications in fundamental physics research, e.g. for the investigation of dark matter. In this context, we present measurements of magnetization by a superconducting quantum interference device (SQUID) and of ¹H nuclear magnetic resonance (NMR) relaxation that highlight Mn₁₂BuAc Single Molecule Magnet as a potential quantum sensor for particle detection. Here we show that the magnetization average relaxation time τ_R , measured both directly by SQUID and indirectly through NMR echo amplitude, is sensitive to even minimal amounts of ionizing radiation impinging on a single crystal. This distinctive sensitivity is enhanced by the metastable character of the chosen initial magnetic state. Theoretical simulations provide an explanation for the shape of the curves describing the evolution of the magnetization towards equilibrium and for the observed behavior of τ_R .

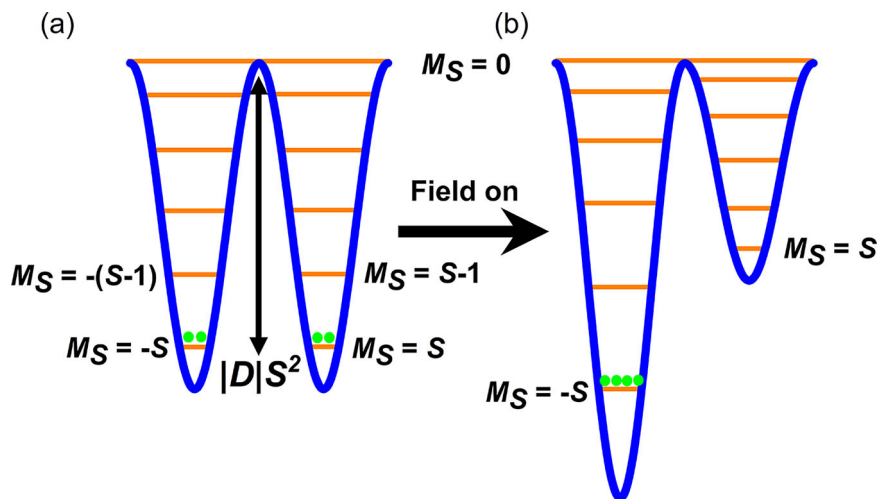
Innovative sensors based on quantum systems and technologies, which intrinsically offer a sensitivity larger than traditional approaches, are expected to provide a boost in the field of fundamental physics¹. The key requirements to use a quantum system as sensor are the presence of discrete levels at well-defined energies, the possibility to control the quantum state and the ability to interact with a given physical quantity of interest, such as an electric or magnetic field². In this respect, molecular nanomagnets appear to be well suited for quantum sensing thanks to their peculiar features, such as chemical specificity and tunability of the electronic structure^{3,4}. These molecules contain mono- or poly-nuclear clusters of transition-metal or lanthanide ions, shielded from one another by surrounding shells of organic ligands. They form crystalline lattices that behave as assemblies of independent, non-interacting units, the quantum behavior of which can manifest at the macroscopic scale^{5–9}. Besides being interesting as testing grounds for fundamental physics, they have been proposed for a variety of technological applications, including high-density information storage^{10–14}, spintronics^{15–19} and quantum computing^{20–23}.

Single Molecule Magnets (SMMs) represent a relevant class among molecular nanomagnets. In SMMs, exchange interactions in the metal core lead to a ground manifold with individual atomic spins locked into a state with large total spin S . When this manifold is subject to an uniaxial magnetic anisotropy of easy-axis type, the $(2S + 1)$ spin sub-levels are split in a double-well potential (Fig. 1a). This energy pattern can give rise to distinctive low-temperature magnetic behavior, such as slow relaxation of the magnetization, driven either by classical Arrhenius-type barrier crossing or by quantum tunneling (QT) through the barrier^{5–8,15,24}. In absence of a transverse anisotropy, the height of the barrier amounts to $\Delta E = |D| S^2$ for integer S and $\Delta E = |D| (S^2 - 1/4)$ for half-integer S , where D is the second-order uniaxial anisotropy constant²⁴. For poly-nuclear systems D depends both on single ion anisotropy (thus on coordination geometry, ligand and spin-orbit coupling of the specific ion) and the exchange coupling pattern²⁵. Without an applied magnetic field, the two wells are equally populated at equilibrium, resulting in zero net magnetization (Fig. 1a). The application of an external magnetic field B along the easy axis of the SMM adds a Zeeman

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Fig. 1 | Double-well energy levels scheme in SMMs. Splitting of the energy levels of a SMM having a ground state spin S and an easy-axis anisotropy in zero field (a), and in presence of an external magnetic field parallel to the easy-axis of magnetization (b). To reach the equilibrium population shown in (b) from that in (a), the spins must overcome the energy barrier. This process is accelerated when two specific levels in the two potential wells align, effectively reducing the barrier height.



interaction term H_Z to the intrinsic SMM Hamiltonian H_{SMM} . This additional term shifts the energy of the two wells in opposite directions and results in unequal equilibrium population and a nonzero magnetization (Fig. 1b). If the molecule is prepared at low temperature T in an initial state with non-equilibrium population of the two wells, reaching equilibrium can require a long time, of the order of hours at $T = 2 \text{ K}$ ²⁴. Indeed, in the absence of effective quantum tunneling pathways, the population transfer from one well to the other one occurs by overcoming the barrier through a phonon-assisted process characterized by a time constant following an Arrhenius law:

$$\tau \approx \tau_0 \exp\left(\frac{\Delta E}{k_B T}\right) \quad (1)$$

where τ_0 is a parameter reflecting the spin-phonon coupling strength, and ΔE is the (effective) barrier height. The value of τ depends on the applied field in a nontrivial manner. A B field modulates, by virtue of Zeeman effect, the axial double-well potential (Fig. 1b), thereby altering the energy barrier that must be overcome for inter-well equilibration. On the other hand, specific values of B enable quantum tunneling pathways, which bypass the classical barrier and thus complicate the simple Arrhenius picture. This occurs when the axial potential of Fig. 1b is such that two specific levels in the two potential wells align. In this situation, small non-axial terms - non-commuting with the axial Hamiltonian - mix the two levels, transforming the crossing into an anticrossing that acts as a tunneling channel through the barrier, effectively decreasing τ ²⁴.

Owing to the exponential dependence of τ on the barrier height and to the possibility to reverse magnetization via quantum tunneling at specific magnetic fields, SMMs provide intriguing opportunities for sensing minute perturbations, such as transverse magnetic fields or incident radiation. Starting from the situation depicted in Fig. 1a ($B = 0 \text{ T}$), and ramping quickly the B field with respect to τ , the stored Zeeman energy is released when reaching the new equilibrium state depicted in Fig. 1b, resulting in a local increase in T . In a non-diluted crystal this can, in principle, induce the relaxation of neighboring molecules and, in some cases (for instance as consequence of a small, localized energy deposit), trigger a magnetic avalanche that rapidly propagates through the entire crystal. This process is analogous to chemical combustion, where energy released in one region drives a reaction in its surroundings. A deflagration occurs when the rate of local energy release is larger than the rate at which energy diffuses away. Magnetic deflagration has been observed in the Mn_{12}Ac ²⁶ and Fe_8 ²⁷ SMMs, as well as in bulk magnets^{28–30}, triggered by proper mechanisms (for instance by surface acoustic waves). In contrast to chemical deflagration, its magnetic analogue is reversible, allowing multiple measurements on the same sample under diverse conditions. In addition, SMMs offer extensive control over the onset of the process. The rate of local energy release is governed by the

relaxation time τ , which can be tuned over several orders of magnitude through temperature variation or by modulating the degree of quantum tunneling (e.g., by tuning the value of the magnetic field along the easy-axis, or adding a small transverse magnetic field component). This effect could be exploited to amplify the response to impinging particles or radiation, enabling their detection³¹. In the framework of a sensing protocol², the spin Hamiltonian can be written as:

$$H_{\text{SPIN}} = H_{\text{SMM}} + H_Z \quad (2)$$

where the Zeeman term $H_Z = \mu_B g \vec{S} \cdot \vec{B}$ acts as a “control” Hamiltonian, while H_{SMM} includes anisotropic and dipolar spin terms (see Eq. (4) below). In our model (see Results and discussion below) the interaction with the impinging radiation is not represented by an additional term in the spin Hamiltonian, as in the case of magnetic field sensing. Instead, the effect of ionizing radiation leads to local heating, ultimately activating phonons both through the action of the radiation and through the local heat wave generated by relaxing spins. In fact, the energy deposited by impinging radiation produces local heating in the irradiated regions. When this triggers spin relaxation from the metastable state, the associated energy release further amplifies the initial signal. Under favorable conditions, this feedback can trigger a magnetic avalanche, observed as a sudden jump in M . However, if the criteria for avalanche ignition are not met - due to insufficient size, geometry, or temperature of the heated region - signatures of signal amplification may still appear. In particular, spins within these locally hotter areas undergo accelerated relaxation dynamics (shorter τ), which can be detected through relaxation measurements.

The possibility to prepare SMMs in a metastable state makes these systems potentially attractive for the detection of small quantities of energy release, particularly those associated with impinging particles or radiation. Given their peculiar energy level pattern, a small quantity of energy absorbed at the anticrossing field can indeed lead to a reversal of magnetization with consequent release of the Zeeman energy and acceleration of the relaxation of the metastable state. The potential of SMMs for this application was investigated in ref. 31, where it was shown that in principle a SMM-based detector could be tuned to be sensitive to $\sim 10^{-3} \text{ eV}$ – 10 eV energy deposits. Such kind of sensors could search for hidden photon dark matter candidates in this energy range, with sensitivities largely exceeding current bounds. A pioneering detection approach reported preliminary magnetometry measurements by means of a Hall sensor, showing jumps in the magnetization in crystals of Mn_{12}Ac SMM attributed to the onset of “magnetic avalanches” triggered by impinging a particles from a low activity source^{32,33}.

In the present work, we investigated the effect of low-activity ionizing radiation sources on crystals of the highly uniaxial $\text{Mn}_{12}^{\text{BuAc}}$, a derivative

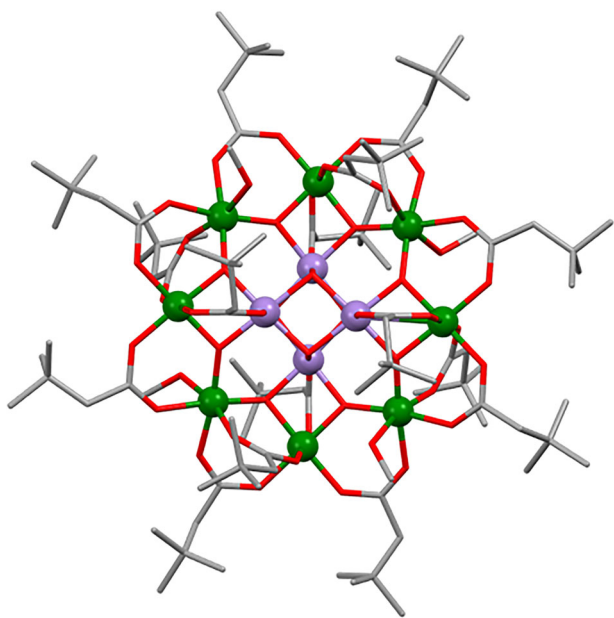


Fig. 2 | Molecular structure of $\text{Mn}_{12}\text{BuAc}$ as seen along the c easy-axis. Mn^{III} ions are reported in green, Mn^{IV} in violet, O in red, and C atoms in pale gray. H ions have been omitted for clarity sake³⁴.

of the archetypal Mn_{12}ac SMM²⁴. Its core (see Fig. 2) is constituted of an internal tetrahedron of four ferromagnetically coupled $\text{Mn}(\text{IV})$ ions (with spin $s = 3/2$) to which an external ring of eight ferromagnetically coupled $\text{Mn}(\text{III})$ ions (with spin $s = 2$) is antiferromagnetically coupled, resulting in a ground state $S = 10$ and an energy barrier of about 60 K. This derivative exhibits reduced disorder and significantly smaller transverse anisotropy than the Mn_{12}ac ³⁴, making it a closer realization of the simple double well model shown in Fig. 1.

We show that, despite not observing the onset of magnetic avalanches under our experimental conditions (where in particular most of the impinging radiations are γ and β , with a marginal α component), exposure to ionizing radiation results in a pronounced acceleration of the spin dynamics. This change far exceeds what would be expected from the small average energy deposited in the sample. These results provide direct evidence for a strong amplification mechanism of the energy released by the ionizing radiation and highlight the potential of SMMs for applications in precision sensing and in searches for faint energy deposits relevant to fundamental-physics experiments.

Results and discussion

Experimental results

Several measuring sessions, using different crystals, were performed with nuclear magnetic resonance (NMR) and superconducting quantum interference device (SQUID) techniques under different experimental conditions. In each measurement campaign the NMR echo signal intensity height $h(t)$ and the magnetization $M(t)$ have been followed for several hours with the two techniques, respectively. The measurement of $h(t)$, being related to the local value of magnetization, is expected to reflect the time evolution of $M(t)$ ³⁵. The experiments were realized with the $\text{Mn}_{12}\text{BuAc}$ crystals facing a semicylinder (SC)-shaped radioactive source and a non-emitting SC, used as a reference, at different B field values (see Methods for details). Figure 3 reports, as an example, the results obtained at $T = 2.2$ K around $B = 0.93$ T (corresponding to a specific anticrossing value). The SQUID measurements acquired at $B = 0.84$ T, which will be in particular considered in our model, are reported in Supplementary Fig. 1. As further reference, the NMR measurements at $B = 0.91$ T are shown in Supplementary Fig. 2.

The data were fitted with stretched exponential functions:

$$h(t) = h_0 + A \left(1 - e^{-\left(\frac{t}{\tau_R}\right)^\beta} \right) \text{ NMR} \quad (3A)$$

$$M(t) = M_0 + M_{\text{sat}} \left(1 - e^{-\left(\frac{t}{\tau_R}\right)^\beta} \right) \text{ SQUID} \quad (3B)$$

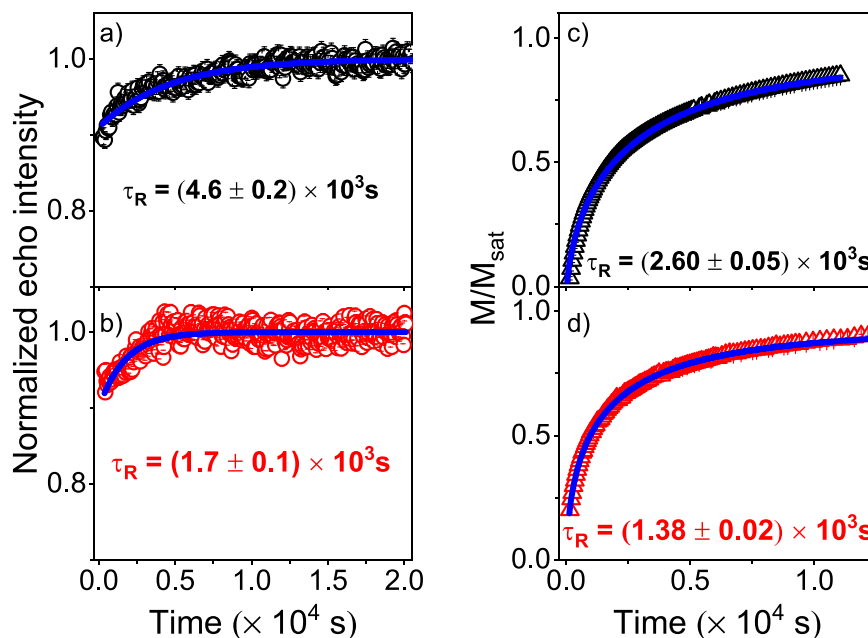
where h_0 is the echo amplitude at $t = 0$ s, becoming $h_0 + A$ at equilibrium ($t \rightarrow \infty$), M_0 is the magnetization at $t = 0$ s (ideally zero), M_{sat} is the saturation magnetization, β is the stretching exponent (typically in the range $0.5 < \beta < 1$) and τ_R is a parameter representing an average relaxation time of the magnetization towards equilibrium. This function is used to interpret the behavior of a wide variety of physical quantities in many different research areas. In particular, it has been found to properly describe the relaxation of the magnetization in SQUID measurements on SMMs, such as Mn_{12}ac ³⁶ and Fe_8 ³⁷, and it is also expected to properly describe the related time evolution of the echo amplitude height. This peculiar behavior is attributed to slight inhomogeneities inside the crystal - leading to locally dependent τ - and to a redistribution of the local fields during the relaxation process, locally affecting the tunneling rate and thus making the relaxation transition probability (and then τ) time dependent. This introduces a deviation from a pure exponential law, which is well described by a stretched exponential. It is worth noting that the shape of the recovery curve is slightly different (*i.e.* different β) in the NMR and SQUID cases. In particular, the fits typically return a $\beta \sim 1$ value for the former measurements (corresponding to a pure exponential trend), which is mainly attributed to the size of the errors affecting the NMR results. Indeed, the NMR data suffered from a high background signal due to both the quite late acquisition of first point of the echo recovery curve (generally not possible before ~ 300 s for experimental limitations) and to the contribution, constant in time, of ^1H NMR echo signal coming from the metallic SC. As further check, we refitted the NMR data at 0.91 T and 0.94 T with Eq. (3A) and a β starting value around 0.55, *i.e.*, the average value obtained by the fits of the SQUID data at similar B values. Despite this, the fit procedure converges to $\beta \sim 1$. By fixing the β value to 0.55, the data are still very well fitted by the new function, giving a different output for the τ_R parameter. We therefore decided to keep β as a free fitting parameter, avoiding assuming a value taken from fits of data obtained with another technique.

The experimental result reported in Fig. 3 is significant in that, for both NMR and SQUID measurements, the best-fits of the data according to Eqs. (3A, 3B) give values for τ_R that are about 2–2.5 times shorter when the crystals are placed in front of the emitting SC with respect to when placed in front of the non-emitting SC. This radiation-induced reduction in τ_R has also been found when repeating the procedure at various B field values around $B = 0.93$ T, as summarized in Fig. 4 and Tables 1 and 2. Similar results have been obtained in other measurement sessions.

Data from a second campaign of measurements performed after about two months on crystals from the same batch as those used to acquire data reported in Fig. 3 are shown in Supplementary Fig. 3 and 4 and Supplementary Table 1 and 2. It can be noticed that a similar reduction in τ_R due to ionizing radiation is still clearly present. However, due to effects that we attribute to possible chemical deterioration of the $\text{Mn}_{12}\text{BuAc}$ crystals over time or to possible differences in crystal alignment with respect to the B field during the mounting in the two campaigns, different τ_R values are obtained for both the non-emitting and emitting configurations at a given reference B field. Moreover, when data taken in the two separate campaigns are rescaled on each other by a scale factor which is obtained at a specific field (0.91 T and 0.93 T for NMR and SQUID, respectively) so to get the same τ_R value, all the results follow an overall consistent trend.

The reduction of τ_R can be explained by assuming that the impinging radiation releases a quantity of thermal energy which locally alters the spin dynamics, inducing a molecular magnetization change in proximity of the

Fig. 3 | Magnetization dynamics around an anticrossing field with and without irradiation. NMR echo intensity vs time, acquired with $B = 0.94$ T at $T = 2.2$ K for the crystals facing the non-emitting (a) and emitting (b) SC; blue lines are fit with Eq. (3A). SQUID recovery curves with $B = 0.93$ T at $T = 2.2$ K, for the crystal in front of the non-emitting (c) and emitting (d) SC; blue lines are fits according to Eq. (3B). All curves are normalized to the equilibrium values obtained by the fit procedures.



QT anticrossing fields (for example due to a facilitated crossing of the barrier and/or to a possible local spin avalanche). This feature is in agreement with theoretical prediction and experimental observation for which the spin avalanche effect occurs with a higher probability in correspondence of the QT fields, thus inducing a shortening of the relaxation time³⁸.

It should be noticed that, given the more local character of the NMR measurements, a comparison between the absolute values of τ_R obtained from the two approaches is not straightforward. This peculiarity of NMR is well known in literature, see e.g.³⁹. Moreover, it should be observed that the presence of several crystals in the NMR setup represents an intrinsic source of disorder. In addition, given the strong temperature and field dependence of the underlying relaxation mechanisms, slight discrepancies may potentially arise from a difference in the actual crystal temperature achieved with the two cryogenic systems and from the slightly different B values considered. However, the observed trend of τ_R vs B is similar for both sets of measurements, with the recovery of the magnetization being faster, as expected, close to the anticrossing value due to QT effects (see e.g. Ref. 6,35,36).

A further indication that the ionizing radiation is affecting the $\text{Mn}_{12}\text{BuAc}$ spin dynamics comes from the observation that the recovery of the magnetization is faster under irradiation in the very first part of data acquisition. Indeed, during the transient time of around 3 minutes required to get a stable B field at the required working value, the irradiated sample systematically recovered about 20% of the saturation magnetization (M_{sat}), while for the non-irradiated sample the recovered M was only a few % of M_{sat} . Again, this difference was systematically found in all SQUID measurements (see Supplementary Fig. 1), also when shielding for α particles. From these observations, when considering the estimated dose rate on the crystals (see Methods below), we can get an upper limit estimate for the sensitivity (minimum detectable energy release) of the crystals of about 6.8 MeV with our approach. This is close to the upper limit in sensitivity of 5.486 MeV reported in ref. 32,33, obtained with the magnetic avalanches approach for a particles absorption.

Regarding the observed stretching exponents $\beta < 1$, these are only marginally affected by radiation and can be primarily attributed to dipolar interactions combined with a small degree of disorder in the spin Hamiltonian⁴⁰ (see next sub-section). To assess whether the observed behavior can be due to the effect of repeated thermal cycles on the $\text{Mn}_{12}\text{BuAc}$ crystals, measurements at B around 0.93 T were repeated on the same sample placed on the non-emitting SC, after heating it to room temperature (several times) and bringing it back to $T = 2.2$ K. The same τ_R

value was found within the experimental error (see e.g. the blue asterisk NMR point in Fig. 4), ruling out this possibility and providing a further confirmation of the reliability and reproducibility of the measurements.

As a final check, we performed a preliminary evaluation of the contribution of the impinging α particles on τ_R , i.e. whether the observed reduction is to be exclusively attributed or not to the α component of the emission from the source. For this scope the crystals were shielded from the α component by covering the emitting SC using a few layers of Teflon® tape. Despite this, the observed τ_R was still significantly shorter than the one observed for the non-emitting SC (Table 2), indicating that the incidence of γ radiation alone is responsible for the effect. Indeed, according to our simulation (see Methods), this component accounts for about 90% of the energy released into the crystals by the source, while the fraction related to the β component is negligible.

Furthermore, we can a priori be confident that the observed effect is not due to a crystal deterioration induced by the radiation. The source is indeed characterized by a very low dose release and its effect is already seen after about 3 minutes of exposition, in correspondence of which there is a relatively low number of absorbed ionizing particles (around 4 α and 200 γ). This hypothesis is confirmed by the comparison of the hysteresis curves before and after about 10 hours of continuous irradiation. As shown in Supplementary Fig. 5, the two hysteresis curves practically coincide, as well as the dM/dB vs B curves, indicating no occurrence of a macroscopic crystal deterioration at least during our measurements time, as expected for the low dose released by the source. A detailed study of the impact of radiation damage on $\text{Mn}_{12}\text{BuAc}$ crystals, to the best of our knowledge hitherto unreported in literature, needs to be performed in the near future and it is beyond the scope of this study.

Model

To model the observed relaxation behavior, we focused on the SQUID measurements at $B = 0.84$ T and 0.93 T. The model was initially calibrated by the “non-emitting” results. The related Hamiltonian^{41,42}

$$H_{\text{SPIN}} = -D\hat{S}_z^2 - C\hat{S}_z^4 + B_4^1\frac{1}{2}(\hat{S}_+^4 + \hat{S}_-^4) - g\mu_B(B + B_{\text{dip}})\hat{S}_z \quad (4)$$

contains quadratic and quartic axial anisotropy terms, and a quartic in-plane anisotropy term. B is the applied magnetic field, with the g -factor $g = 2$. The axial terms determine the values of the level-crossing fields, while the quartic transverse term introduces small splitting at selected crossings,

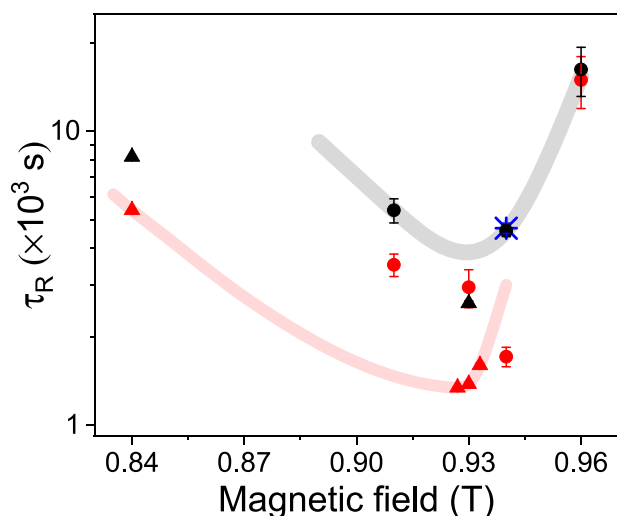


Fig. 4 | Average relaxation time τ_R as a function of B . The τ_R values are extracted from the fits of the NMR (circles) and SQUID (triangles) data acquired at various B fields around $B = 0.93$ T and $T = 2.2$ K, with the crystals facing the non-emitting (black) and emitting (red) SC. The error bars (visible only when exceeding the symbol size) represent the uncertainty on the τ_R values from the fitting procedure. The blue asterisk is the NMR measurement repeated after heating up to room temperature. The gray-shaded (pink-shaded) bands are guides for the eye, related to NMR non-emitting (SQUID emitting) data.

thereby opening tunneling pathways. B_{dip} accounts for dipolar interactions at the mean-field level. Although small, they can produce substantial effects in the time-dependent magnetization $M(t)$ close to crossing field. In fact, the dipolar mean-field is itself time-dependent, $B_{dip} \propto M(t)$, so that the total field $B + B_{dip}$ felt by the spins sweeps across the anticrossing region⁴³, leading to a pronounced time dependence of the relaxation rate and an overall non-exponential relaxation⁴⁴. We take $B_{dip} = 0.025M(t)/M_{sat}$ T, corresponding to a maximum dipolar field of 0.025 T. This value is roughly half that found in $Mn_{12}ac$ for elongated samples^{43,45}, consistent with the larger unit-cell volume of $Mn_{12}BuAc$.

Magnetic relaxation is driven by spin-phonon coupling, which we model using a widely adopted simplified approach. This assumes that the coupling arises from small rotations of the rigid molecule around its equilibrium position^{46–49}. In this framework, phonons modulate the orientation of the anisotropy axes without altering the parameters D , C and B_4^1 . Combined with the assumption of isotropic Debye phonons, this choice is particularly convenient, as it provides a univocal form for the coupling. The first-order spin-phonon coupling contribution is

$$H_{SP,rot} = \sum_{\alpha=x,y,z} \sum_{k,\lambda} i[H_{SPIN}, S_{\alpha}] \gamma_{\alpha,k,\lambda} (a_{k,\lambda}^{\dagger} + a_{-k,\lambda}) \quad (5)$$

where $a_{k,\lambda}^{\dagger}$ creates a phonon of wavevector k in branch λ , and the coupling strength $\gamma_{\alpha,k,\lambda}$ depends on phonon frequencies and polarizations and can be calculated from density and sound velocities. This form of the magnetoelectric coupling neglects both the internal distortions of the molecule - modeled as rigid - and the actual non-Debye characteristics of phonons. These factors can introduce corrections to the rotational model, especially given the relatively large spectral energy scale, on the order of tens of meV. To account for the limitations of this approximation in the most economical way, we treat the overall energy scale in Eq. (5) as a free parameter⁴⁹. Then, the rate for the transition from level m to level n of H_{SPIN} is

$$W_{mn} = A\Delta^3 (e^{\frac{\Delta}{kT}} - 1)^{-1} \sum_{\alpha=x,y,z} |\langle m | [H_{SPIN}, S_{\alpha}] | n \rangle|^2 \quad (6)$$

Table 1 | Average relaxation time τ_R from NMR measurements

B (T)	Configuration	τ_R (s) $\times 10^3$
0.91	Non-emitting	5.4(5)
	Emitting	2.6(3)
0.94	Non-emitting	4.6(2)
	Non-emitting*	4.7(2)
0.96	Emitting	1.7(2)
	Non-emitting	16(3)
0.96	Emitting	15(3)

Values of the τ_R parameter extracted from the fits - according to Eq. (3A) - of the NMR data, taken at different B fields around $B = 0.93$ T and $T = 2.2$ K. The β values are very close to 1. The error on the parameter is reported in parenthesis.

* after thermal cycle.

Table 2 | Average relaxation time τ_R from SQUID measurements

B (T)	Configuration	τ_R (s) $\times 10^3$	β
0.930	Non-emitting	2.60(5)	0.60(1)
	Emitting	1.38(2)	0.519(2)
0.927	Emitting	1.34(2)	0.511(2)
	α shielded	1.77(3)	0.69(1)
0.933	Emitting	1.60(2)	0.522(2)
	α shielded	1.93(4)	0.69(1)
0.840	Non-emitting	8.19(4)	0.811(5)
	Emitting	5.39(1)	0.703(2)
0.840	Emitting	5.78(3)	0.602(2)
	α shielded	5.78(3)	0.602(2)

Values of the τ_R and β parameters extracted from the fits - according to Eq. (3B) - of the SQUID data, taken at different fields around $B = 0.93$ T and $T = 2.2$ K. The error on the parameters is reported in parenthesis.

where $\Delta = E_n - E_m$ and A is the free scale parameter. For a fixed Zeeman contribution in Eq. (4), the associated master equations for the time-dependent state populations of the 21 spin states yield 20 distinct time constants for each temperature and field. Among them, the longest, τ , corresponds to the equilibration between the two potential wells via barrier crossing, and thus governs the long-term relaxation of magnetization. This well-established behavior and the way τ depends on the B field, are independent of the fine details of the spin-Hamiltonian (Eq. (4)) and the spin-phonon coupling (Eqs. (5) and (6)). Steps in the hysteresis curves are associated with minima in the field-dependence of τ , which occur when a tunneling pathway opens in correspondence with level crossings. Note, however, that for relaxation measurements with the present protocol the dipolar contribution to the field, which grows in time, makes the rates in Eq. (6) time dependent; consequently, the corresponding τ is also time dependent and spans a range of values during the measurement. In general, a stretched exponential curve (see Eqs. 3A and 3B) fits the resulting $M(t)$ very well, yielding values of τ_R and β parameters that can be compared with experiments.

A detailed model of the effect of impinging radiation is out of reach due to the complexity of the associated processes. It is here assumed that the related energy deposit is producing a local heating in tiny regions of the crystal throughout the relaxation measurement. The shape, extension and heating of these regions is likely to be highly irregular and variable. The elevated temperature in these regions accelerates relaxation from the metastable state, while the associated energy release will amplify the effect by heating adjacent volumes of the crystal. The heat wave propagates according to a diffusion equation with a source term, potentially leading to a global avalanche³¹, with the initial hot region acting as the seed. This scenario might

lead to sudden magnetization jumps, which have been reported to occur with α irradiation^{32,33}, but that are not observed with our experimental configuration, characterized by a different irradiation geometry and composition (γ dominated, see Methods for a detailed characterization of the radioactive SC) and by a higher T . Nevertheless, even in absence of avalanche effects, the local reduction of τ_R may decrease the spatially averaged τ_R of the crystal, an effect that can become detectable if the heated region is sufficiently large.

Since a microscopic description of the hot regions is prohibitively complex, we adopt a simplified approach to characterize, in coarse terms, the observed reduction in the relaxation time. We assume that the sample contains two components, each described by master equations with rates of the form (6). One component is at the thermostat temperature $T = 2.2$ K, while the second one is characterized by a higher effective temperature (T_H). The number of spins in the hot component is taken as a fixed fraction (w_H) of the non-relaxed spins. The underlying idea is that most of the heat forming the hot region is generated by the decay of metastable spins initially triggered by an impinging ionizing particle. Thus, its volume is expected to be time-dependent, tracking the fraction of metastable spins: it is maximal at $t = 0$ s and decreases to zero as $M(t)$ approaches saturation. The weight w_H and temperature T_H are treated as free parameters, as our sample-averaged experimental information is insufficient to determine both uniquely, and comparably good fits can be obtained by compensating changes in w_H with opposite changes in T_H .

If B_{dip} is neglected in Eq. (4), simulations by master equations produce, as expected, mono-exponential relaxation of M . As already mentioned, the stretching exponent $\beta \neq 1$ in Eqs. (3A–3B) suggests that τ has a distribution of values. This is consistent with β being smaller at $B = 0.93$ T, in the region of strong tunneling, where τ is particularly sensitive to details in Eq. (4), such as the dipolar contribution B_{dip} . The solid black line in Fig. 5 shows the calculated B -dependence of the τ_R parameter which fits well the two measured values in the absence of radiation. The calculation predicts a pronounced stretching at $B = 0.93$ T, with an exponent β close to 0.7, arising from the time-dependent B_{dip} . In contrast, for $B = 0.84$ T the calculated stretching is negligible.

For the measurements performed under irradiation we simulate $M(t)$ using the two-component model described above and extracting the corresponding τ_R . The entity of the observed reduction of τ_R is well captured by assuming $T_H = 2.8$ K and $w_H = 1.4\%$, but similar results are obtained with slightly larger (smaller) T_H and correspondingly smaller (larger) w_H .

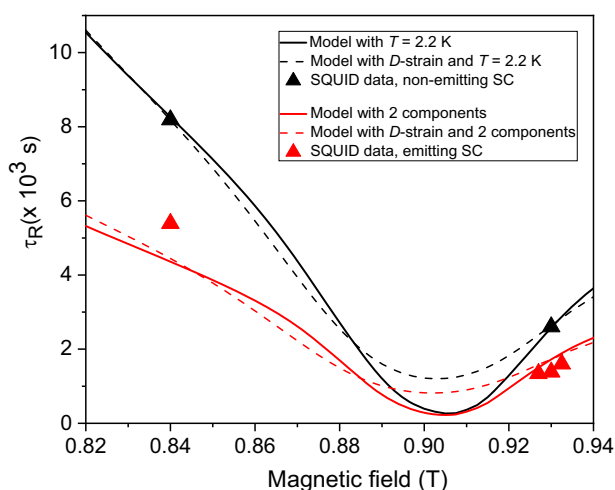


Fig. 5 | Simulated average relaxation time τ_R vs applied magnetic field. Solid black line: calculation from Eqs. (4) and (6), with $T = 2.2$ K, $D/k_B = 0.57$ K (about 1% larger than in ref. 41), $C/k_B = 1.2$ mK⁴¹, $B_d^4/k_B = 6.2$ μ K⁴², $A = 1.2$ meV⁻⁵ s⁻¹ and $B_{dip} = 0.025M(t)/M_{sat}$ T. Dashed black line: same calculation including Gaussian D -strain with width $\sigma_D = 0.02$ D , and $A = 1$ meV⁻⁵ s⁻¹. Red lines: same calculations including a second component, with $T_H = 2.8$ K and $w_H = 1.4\%$.

The measured value $\beta \cong 0.8$ at $B = 0.84$ T in the absence of irradiation indicates additional sources of non-exponential behavior beyond the dipolar interactions considered so far. These are most likely associated with slight disorder in the parameters of Eq. (4), which is commonly observed in molecular nanomagnets. For example, ref. 40 reports a distribution of D (D -strain) with a width of 1% for the Fe₈ nanomagnet. In Mn₁₂ac, a D -strain of approximately 2% has been observed, along with a distribution of the g -factor (g -strain) of 0.8%. Similar effects may also be expected for the tunneling parameter B_d^4 or for second-order tunneling terms such as B_2^2 , which can be activated by perturbations of the tetragonal point symmetry. Clearly, it is impossible to account for all such fine details within a model calculation. As an illustration, we consider the effect of D -strain by assuming Gaussian-distributed D values centered at $D_0 = 0.57$ K with a width $\sigma_D = 0.02$ D_0 . As shown in Fig. 5, the minimum in the field dependence is smoothed out by the resulting distribution of tunneling fields. In addition, the stretched exponential behavior is found over the whole field range, with $\beta \approx 0.9$ at $B = 0.84$ T. Different values of β can result from different assumptions on the type of disorder. However our purpose is not to fit such fine details, but just to show that these do not alter our conclusions as the difference between black and red curves is not washed out.

It is worth noting that the presented model has been tuned only on the SQUID measurements acquired at $B = 0.84$ T and 0.93 T. Anyway, when considering data from the second measurement campaign, once properly rescaled (as previously reported in Experimental results), the τ_R values are still reasonably fit by the model tuned with the first data campaign (see Supplementary Fig. 6). This observation is a further verification of the confidence of the model.

About the observed discrepancies, it should be noticed that a global B -independent rescaling factor could a priori not be adequate to account for the possible effects originating the differences in τ_R observed in the two campaigns.

With regards to the NMR results, the associated values of τ_R cannot be quantitatively fit by the same model fitting SQUID data. Although NMR also shows a comparable acceleration of the relaxation under irradiation, the associated τ_R are generally longer than expected. As already discussed above, small differences in the actual crystal temperatures between the two cryogenic setups (on the order of 0.1 K) or in the B field values, together with the use of multiple crystals in the NMR measurements, are likely responsible for this difference.

By using the dose rates estimated from simulation (see Methods), it was possible to obtain - from relevant properties of Mn₁₂BuAc⁵⁰ - an estimate of the crystal temperature increase due to impinging ionizing radiation during the measurements. A T increase of about 10^{-4} K over a typical measurement period of 4 h is expected in the non-realistic experimental hypothesis that the cryostat is not properly refrigerating the crystal during the long-time of the measurement. This clearly represents an overestimated upper limit and, given the typical performances of the cooling systems, we can reasonably be confident that there is not substantial global thermal effect due to impinging radiation. This is confirmed by the fact that in a simplified “single” component implementation of the theoretical model, a global thermal increase of the order of 0.05 K is needed to simulate reasonably well the measured recovery curves for the irradiated crystal, as well as the τ_R extracted from the corresponding fits.

An alternative explanation is that the seed of the hot region is not primarily thermal in origin. Ionizing radiation may produce localized structural perturbations or microscopic damage in the region of the particle energy release, which could modify the spin Hamiltonian. In particular, disorder affecting the transverse anisotropy terms may reduce the effective barrier height enhancing tunneling probabilities, thereby shortening the local relaxation time without invoking a significant temperature increase. Although we cannot resolve such effects directly, their presence cannot be excluded and may contribute to the observed acceleration of the relaxation. However, it is worth to stress that the experimental results of the current study give no evidence of such kind of effects (see Experimental results and Methods). From this perspective, further dedicated experimental studies are

required in order to investigate the impact of radiation damage on SMMs crystals.

Conclusions

In conclusion, the dynamical properties of $\text{Mn}_{12}\text{BuAc}$ SMM crystals were studied by means of NMR relaxation and SQUID magnetometry under the effect, or not, of a low-activity ionizing radiation source. At $T = 2.2$ K, exposure to radiation accelerates the relaxation of the magnetization by a factor of 2–2.5. This effect is far from what would be expected as a consequence of the small average energy deposited in the sample. Theoretical modeling based on the giant-spin Hamiltonian, dipolar mean-field effects, and phonon-assisted relaxation explains the field dependence and stretching of $M(t)$ in the absence of radiation. To account for the ionizing radiation-induced acceleration, we introduced a minimal two-component description in which a small fraction of spins temporarily experiences a higher local temperature. This phenomenological approach captures the essential physics of the effect: the amplification mechanism arising from the feedback between local heating and the release of Zeeman energy during relaxation. While this heating remains confined - preventing the onset of a macroscopic magnetic avalanche - it is sufficient to alter the average relaxation rate of the sample.

These results provide a proof of principle that SMM crystals can transduce very small, spatially localized energy depositions into measurable changes of their relaxation dynamics. Importantly, the effect is observed for radiation largely dominated by low energy (below 100 keV) photons, which apparently are not able to trigger avalanches at least in our experimental setups. This shows that SMM-based quantum sensors do not necessarily need to rely on macroscopic magnetization jumps and can instead exploit subtler dynamical signatures. The sensitivity demonstrated here (with an upper limit estimate of about 6.8 MeV), together with the intrinsic tunability and reproducibility of SMMs, highlights their potential for applications in precision sensing and in searches for faint energy deposits relevant to fundamental-physics experiments.

Further experimental and theoretical investigations - especially those addressing microscopic heat transport and radiation-spin coupling - will be essential to optimize and fully exploit this sensing paradigm and so to

characterize SMMs as particle detectors. In particular, dedicated and systematic studies will be required to properly understand the separate impact of α , β and γ ionizing radiation on the observed reduction of the average relaxation time τ_R .

Methods

Polycrystalline samples of $\text{Mn}_{12}\text{BuAc}$ were prepared according to published procedure³⁴. Crystals with typical dimensions up to $3 \text{ mm} \times 0.5 \text{ mm} \times 0.5 \text{ mm}$ (corresponding to a mass of $\sim 1 \text{ mg}$) were obtained. Their quality was assessed by X-ray diffraction analysis and confirmed by studying the hysteresis loop with a SQUID magnetometer, showing the well-known shape with steps associated to an enhanced relaxation due to quantum tunneling at the expected anticrossing B values (see Supplementary Fig. 7). This figure also evidences that the overall shape of the hysteresis curve (i.e. anticrossing B values) does not change for irradiated crystals. This indicates that (at least under the low dose rates of our source) the irradiation does not produce any relevant damage capable of changing the energy level structure of the molecular cluster.

Semicylinder (SC)-shaped radioactive sources were obtained by mechanical processing of special welding electrodes - composed by 98% W and 2% Th - to fit with the experimental setup needs. The presence of Th and of its decay products makes the SC a very low activity source with simultaneous emission of α , β and γ ionizing radiations. A precise source emission characterization was performed at the Joint Research Centre in Ispra⁵¹, giving the surface radiation fluxes and energy ranges summarized in Table 3. In order to perform reference measurements under the same experimental conditions, but in absence of impinging radiation, non-emitting SCs - made of 98% W and 2% La - were used.

Either a single crystal or few crystals (covering an area of $\sim 3 \text{ mm} \times 4 \text{ mm}$) of $\text{Mn}_{12}\text{BuAc}$ were used in the SQUID and in the NMR measurement, respectively. Each crystal was glued on a polyester film (Mylar[®]) strip and mounted in front of the SC so to have its lateral surface being directly irradiated at a distance of approximately 0.5 mm (see Fig. 6). The same experimental setup was used for both the non-emitting and emitting configurations.

A dedicated simulation of source and sensor materials, as well as of the experimental configuration, has been performed using the PHITS (Particle and Heavy Ion Transport code System)⁵² version 3.34 code. This study allowed to get more insight about the details of our source emission and of the related energy absorption in the $\text{Mn}_{12}\text{BuAc}$ crystals. According to Table 3, it has been found, in particular, that the absorbed dose is mostly driven by γ radiation: the photons release in the crystal an average energy which is about a factor of 10 greater than the one released by α particles, while the β release results negligible. The photon simulated energy spectrum is shown in Supplementary Fig. 8. From this spectrum, a correction factor of about 1.3 has been derived for the γ surface flux, to account for the fraction of low energy photons ($E_\gamma < 59 \text{ keV}$) not detected in the experimental characterization of the source. As a result, we finally estimated a dose rate on a single crystal of about $3 \times 10^{-8} \text{ Gy/min}$ and $3.6 \times 10^{-7} \text{ Gy/min}$ due to α and γ particles, respectively. Furthermore, being highly ionizing, α particles deposit their energy inside the crystal only within a few tens of μm from the surface, whereas the γ absorption exhibits a typical exponential trend.

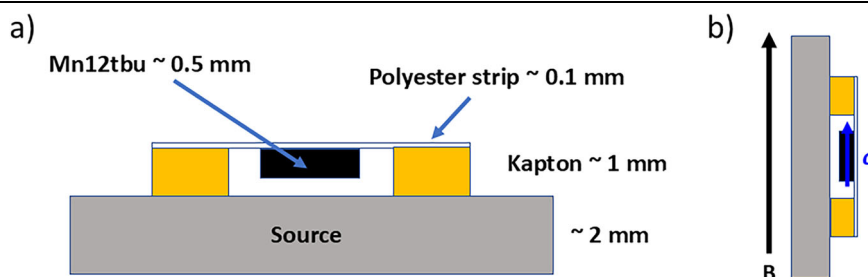
Table 3 | Characterization of ionizing radiations emitted from the radioactive sources

	α emission	β emission	γ emission
Surface flux (particles/ $\text{mm}^2 \cdot \text{min}$)	~ 0.9	~ 26	~ 268
Energy range	(0–9.8) MeV	(0–834) keV	(59–1954) keV
Average dose (Gy/particle)	$(2.2 \pm 0.3) \times 10^{-8}$	negligible	$(9.0 \pm 0.2) \times 10^{-10}$

Top and middle rows: measured surface radiation fluxes and energy range. Bottom row: simulated average radiation dose per particle in the crystal volume. The γ surface flux has been corrected by a factor of about 1.3, derived from simulation, to account for the fraction of low energy photons ($E_\gamma < 59 \text{ keV}$) not detected in the experimental characterization of the source.

Fig. 6 | Scheme of the SQUID sample mounting.

a The relative position of the $\text{Mn}_{12}\text{BuAc}$ crystal with respect to the source is evidenced. **b** Schematic view of the crystal inside the SQUID. The magnetic field B is applied parallel to the c easy axis of the crystal. A similar setup has been used for the NMR measurements.



Finally, it has been found that mainly low energy photons ($E < 100$ keV, absorbed by the dominant photoelectric effect) interact, while higher energy photons ($E > 100$ keV, where they mainly interact by Compton effect and at higher energies also by pair production) rarely interact. More into details, around 44% of the photons in the low energy range, representing about 40% of the total γ emission, are absorbed, while most (by far more than 99%) of the photons in the higher energy range cross the crystal without interacting.

The ^1H -NMR Hahn echo measurements were carried out utilizing a phase-coherent spin-echo pulse spectrometer Apollo by Tecmag (Houston, USA). The echo was generated by a $\pi/2 - \pi$ pulse sequence with a typical $\pi/2$ pulse length of about 2 μs . The dc magnetization measurements were performed by a superconducting quantum interference device (SQUID) magnetometer MPMS-XL7 by Quantum Design (San Diego, USA). The two techniques shared the same measurement protocol consisting of: zero field cooling the sample until reaching $T = 2.2$ K; ramping B (parallel to the crystal easy axis) up to the desired value; acquiring the signal for hours in order to follow the recovery of M . It is worth noting that also in NMR this procedure allows to measure the relaxation time τ_R of Mn_{12} - $^1\text{BuAc}$, since the proton NMR echo intensity changes in time when the measurement is done off equilibrium, i.e. during the recovery of the crystal magnetization toward equilibrium^{54,53}. The NMR and SQUID measurements were acquired at different B values around 0.93 T, at which a jump in the hysteresis loop is observed (see Supplementary Fig. 7) in agreement with what reported in literature^{6,41,53}. The choice of this field, at which two specific levels in the two potential wells align enabling the onset of QT, was driven by the need to have a NMR echo signal sufficiently high.

Data availability

Experimental data are provided with this paper in Supplementary Data 1.

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Author contributions

M.F., A.L., G.L., P.S., M.Ma. and L.S. conceived the research. F.S. synthesized the materials and grew the crystals. A.C., M.F., G.L. and L.S. carried out the SQUID measurements. F.B., A.L. and M.Ma. performed the NMR measurements. E.G. and M.Me. characterized the radioactive sources. S.F. simulated the interaction of the radiation with the crystal. A.C., M.F., A.L., G.L., M.Ma. and L.S. performed the data analysis. P.S. developed the theoretical model and performed the simulations. All authors critically followed each aspect of the research and contributed to drafting and editing the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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