

Chapter 1

From Quantum Relaxation to Resonant Spin Tunneling

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Abstract A brief historic review of the research on quantum tunneling of magnetization is given and the story behind the discovery of resonant spin tunneling in Mn-12 acetate is told with the emphasis on the underlying physics. Important areas of studies in this field are mentioned and thoughts are presented on the future of research on molecular magnetism.

1.1 Historic Notes

My interest in quantum tunneling of magnetization was instigated by the 1988 Physical Review Letter of Chudnovsky and Gunther [1], in which they proposed that superparamagnetic behavior of small magnetic particles may not freeze down to absolute zero due to quantum tunneling of the magnetic moment. In 1990 Chudnovsky came to Barcelona on my invitation and we discussed at length possible experiments on quantum tunneling of the magnetization. I chose to study magnetic relaxation (which is sometimes called magnetic after-effect) in arrays of magnetic particles and other magnetic systems and to see if it persists down to very low temperature and eventually becomes independent of temperature when quantum effects take over thermal fluctuations.

From 1990 to 1995 we mounted a large effort at the University of Barcelona to observe these effects in systems of small particles, random magnets, and superconductors. This effort was reviewed in the book of Chudnovsky and myself “Macroscopic Quantum Tunneling of Magnetization” (Cambridge University Press, 1998) [2]. Published experimental results demonstrated that non-thermal magnetic relaxation was, indeed, present in all of the above systems. Moreover, theoretical estimates of the temperature of the crossover from thermal to quantum relaxation agreed well with our findings. Similar results on small particles were obtained by Berkowitz at the University of California—San Diego [3]. Barbara at CNRS-Grenoble [4], O’Shea at the University of Kansas [5, 6], and Arnaud as at the University of Zaragoza [7] observed non-thermal magnetic relaxation in bulk materials

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as well. These findings created a significant boost for research on quantum tunneling of magnetization that culminated in our discovery of resonant spin tunneling in Mn-12 acetate [8].

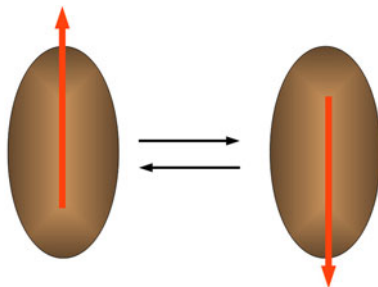
The discovery of magnetic bistability of Mn-12 by Sessoli et al. [9] in early 1990s made us think about definitive experiments on this system that would prove the existence of spin 10 tunneling beyond a reasonable doubt. The beauty of Mn-12, as compared, to systems of small particles, was in that the molecules, unlike magnetic particles, were identical, so that the tunneling rates had to be nearly the same for a macroscopic number of molecules in a solid. This implied that the decay of the magnetization of the Mn-12 sample would be similar to nuclear decay, that is, exponential in time. (This turned out later to be only approximately true because of the distribution of dipolar and nuclear hyperfine fields, as well as due to solvent disorder inside individual molecules and due to crystal defects.)

In 1995 I came to New York for three months on the invitation of Chudnovsky to do magnetic measurements with the group of Myriam Sarachik at City College. They had just bought a Quantum Design Magnetometer that they were not sure how to use. Our goal was to observe magnetic tunneling at low temperature. Previously, Chudnovsky and I had chosen to work on Mn-12. To have samples of Mn-12 I invited Ron Ziolo, head chemist at Xerox Corporation, with whom I had extensive previous collaboration on small particles, to join our group. I was also pleased to work with Jonathan Friedman, a talented Ph.D. student of Prof. Sarachik. Jonathan came up with a brilliant idea to align microcrystals of Mn-12 (that we had at the time) in a styacast using a high magnetic field. As far as magnetic measurements were concerned, this made the array of microcrystals equivalent to a large single crystal and allowed us to observe, for the first time, the equidistant steps in the magnetization curve of Mn-12.

Our findings and their correct explanation in terms of resonant spin tunneling, were first reported and publicly discussed at the MMM conference in Pittsburgh in 1995. Same year we submitted a paper to Physical Review Letters that was published [8] in May 1996. During spring of that year my group at the University of Barcelona, together with the group of Juan Bartolome at the University of Zaragoza, confirmed equidistant tunneling resonances in measurements of the ac susceptibility of Mn-12 [10]. Barbara's group reported in a follow-up article [11] experiments performed on single crystals which corroborated these results.

From 1996 research on spin tunneling in molecular magnets picked up. Experimental landmarks worth mentioning are topological interference in spin tunneling demonstrated by Wernsdorfer and Sessoli [12], determination of the tunneling terms in the spin Hamiltonian of Mn-12 by the groups of Kent and Hill [13], and the works of New York and our groups on magnetic deflagration [14, 15]. There also has been a remarkable effort by chemists to synthesize new molecular magnets (Christou, Hendrickson, and others) with hundreds of them now available. With low temperature physics suffering from the rising cost of helium the future of this field depends, in my opinion, on whether research on molecular magnets finds any applications.

Fig. 1.1 Quantum tunneling of the magnetic moment in a single-domain particle



1.2 Early Experiments on Magnetic Tunneling at the University of Barcelona

In the absence of the magnetic field, single-domain magnetic particles have two opposite directions of the magnetic moment that correspond to the minimum of the magnetic energy. This is true independently of the shape or other anisotropic factors and is a consequence of the time-reversal symmetry. Magnetic field breaks time-reversal symmetry, making the two minima different in energy. Still, under a certain field strength, there is an energy barrier between the two states. It is due to the combined effect of shape and magneto-crystalline anisotropy of the particle. In large particles the barrier is high and the magnetic moment of the particle is frozen in a certain direction. In smaller particles, thermal fluctuations may kick the magnetic moment over the barrier, leading to the phenomenon of superparamagnetism. As temperature goes down, thermal fluctuations die out and magnetic moments of even the smallest particles would be frozen in the absence of quantum transitions. If, however, there were quantum underbarrier transitions (tunneling) between energy minima, the superparamagnetism would persist down to absolute zero, see Fig. 1.1.

Similar effects must exist in bulk magnetic materials where the change of the magnetization occurs through motion of domain walls. The barriers for such a motion are provided by pinning of the domain walls by defects in a solid. In the absence of the external magnetic field, the lowest energy state of a macroscopic magnet has zero total magnetic moment owing to the breakage into magnetic domains of opposite magnetization. Motion of domain walls separating magnetic domains is required to achieve this lowest energy state. At high temperature, thermal fluctuations kick the domain walls out of potential wells created by the pinning rather effectively, thus providing good mobility of the walls. Consequently, a permanent magnet would lose its magnetic moment rather fast. At low temperature, however, thermal processes slow down and the only reason for domain walls to escape potential wells created by the pinning would be quantum tunneling. This also applies to type-II superconductors, where change in the magnetization requires motion of Abrikosov flux lines pinned by defects.

The earliest mentioning of the possibility of quantum tunneling of the magnetic moment, probably, belongs to Bean and Livingston [16] who noticed that relaxation of the magnetization in systems of small particles did not disappear completely

down to very low temperature. Later, various authors observed similar effect in bulk magnetic materials and superconductors, and speculated about tunneling of domain walls and Abrikosov flux lines [2]. Sustained progress in this direction was impeded by the absence of the general theory of such phenomena. Chudnovsky in 1979 noticed [17] that quantum tunneling of magnetization is given by the imaginary time solutions (instantons) of the Landau-Lifshitz equation that had been traditionally used to describe classical micromagnetic phenomena. This suggestion received further development in several papers published by van Hemmen and Sütö [18], Enz and Schilling [19], and Chudnovsky and Gunther [1, 20] between 1986 and 1988, the latter two papers being a major breakthrough in the understanding of the tunneling of single domain particles at low temperatures. In the first of these papers they computed the WKB exponent and estimated the temperature of the crossover from thermal to quantum superparamagnetic behavior for single-domain magnetic particles. The second paper dealt with quantum nucleation of domains in a magnetic film. These papers triggered modern interest, including my own, to quantum tunneling of magnetization.

The most important theoretical prediction for experiment was that the temperature of the crossover from classical to quantum regime scaled with the components of the magnetic anisotropy for both, uniform under-barrier rotation of the magnetic moment in small particles [1] and quantum diffusion of domain walls in bulk materials [20]. The challenge for experiment was to test this and other predictions in the presence of the broad distribution of energy barriers in systems of small particles and bulk magnetic materials. Exponential dependence of the tunneling rate on the barrier height stretches the lifetimes of metastable magnetic states from nanoseconds to the lifetime of the Universe. This, in fact, is a general situation in nature whether one studies magnetic relaxation or relaxation of the elastic stress in solids. The main point is that only those barrier heights, U , which have associated a flipping time of the order of the experimental time scale t , contribute to the relaxation process towards the global thermodynamic equilibrium. That is, measuring the slow evolution of magnetization with time allows us to sweep the barrier height distribution: the bigger the time is, the higher the barriers are.

Observation that allows one to investigate such situation quantitatively is that only certain barrier heights, U , contribute to the relaxation at a time t that elapsed since the preparation of the material.

The rate of thermal decay of the metastable states with a barrier U at a temperature T is given by

$$\Gamma_U = \nu \exp\left(-\frac{U}{k_B T}\right) \quad (1.1)$$

where ν is some pre-exponential factor (the attempt frequency) and k_B is the Boltzmann constant. The characteristic time of the decay depends exponentially on the barrier height: $\tau_U = 1/\Gamma_U = \tau_0 \exp(U/k_B T)$ where τ_0 is the so-called microscopic attempt time. The barriers that contribute to the relaxation at a time t are determined by the equation $t = \tau_U$, which gives $U = k_B T \ln(t/\tau_0)$. By the time t metastable states that have lower barriers have already decayed, while metastable states with

Fig. 1.2 Logarithmic relaxation of the magnetic moment of the array of CoFe_2O_4 nanoparticles measured at various temperatures [21]

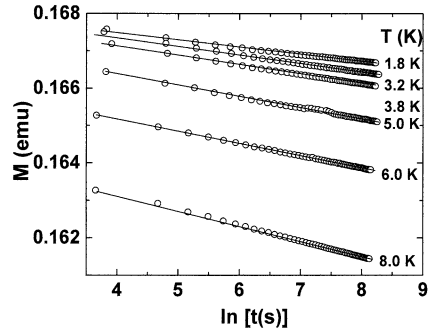
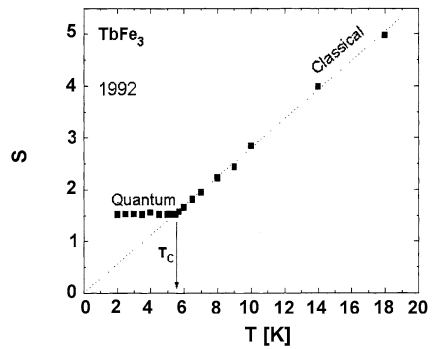


Fig. 1.3 Temperature dependence of the magnetic viscosity of TbFe_3 nanoclusters (1993) [22]

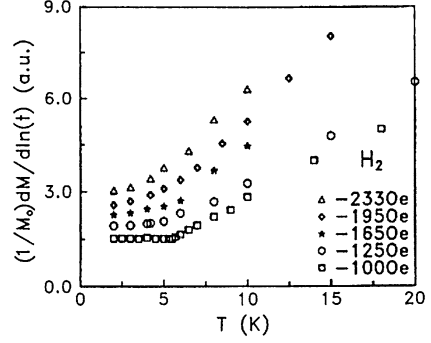


higher barriers have not decayed yet. Analysis of this situation [2] shows that the change in the total magnetization, M , depends linearly on $T \ln(t)$. Such logarithmic relaxation is known in many systems, magnetic or non-magnetic, and is related (through Fourier transform) to the notorious $1/f$ noise. An example of the logarithmic time relaxation in a system of CoFe_2O_4 nanoparticles [21] is shown in Fig. 1.2.

Thermal logarithmic relaxation implies that the derivative $dM/d \ln t$ (called magnetic viscosity) is proportional to temperature and must go to zero as temperature is lowered. Failure of the magnetic viscosity to go to zero in the limit $T \rightarrow 0$ would be an indication of non-thermal magnetic relaxation. This was the basis of our early experiments on quantum tunneling of magnetization. Temperature dependence of the magnetic viscosity in a system of TbFe_3 nanoclusters [22] is illustrated in Fig. 1.3. It clearly shows the plateau in the magnetic viscosity below 6 K, in agreement with theoretical expectation for quantum tunneling in these clusters having very high magnetic anisotropy. Another theoretical prediction is that the crossover temperature goes down with the decreased energy barrier. The latter can be achieved by applying the magnetic field since the field of a certain strength destroys anisotropy barriers altogether. The observed decrease of the crossover temperature on the magnetic field in TbFe_3 nanoclusters is shown in Fig. 1.4.

In the early 1990s similar results were obtained by us and other groups in various systems, including small particles, mesoscopic wires, thin films, bulk magnetic materials, as well as type-II superconductors [2]. Tunneling in antiferromagnetic clus-

Fig. 1.4 Field dependence of the low-temperature magnetic viscosity of TbFe₃ nanoclusters (1993) [22]



ters has been studied (Chudnovsky and Barbara, Awschalom, Tejada). All data for magnetic materials were combined by us in a table showing the dependence of the crossover temperature on the strength of the magnetic anisotropy. It proved correlation between the two quantities, with a clear tendency of the crossover temperature being proportional to the anisotropy [2].

As years went by, measurements of individual nanoparticles had become possible. Wolfgang Wernsdorfer pioneered such measurements in Grenoble. In 1997 he reported [23] evidence of non-thermal magnetic relaxation in a single magnetic nanoparticle of barium ferrite below 1 K. The reduction of the energy barrier needed to provide a significant tunneling rate was achieved in Wernsdorfer's experiment by application of the magnetic field that was close to the field destroying the barrier.

1.3 Experiments on Mn-12

In the early 1990s Roberta Sessoli from Gatteschi's group in Florence discovered [24] that Mn-12 acetate molecules had a 65 K barrier between two lowest energy states with opposite directions of spin $S = 10$. Consequently, a crystal of Mn-12 molecules was equivalent to a system of identical superparamagnetic particles. This removed the challenge of broad barrier distribution that clouded interpretation of experiments with magnetic particles and placed Mn-12 at the top of candidates for observation of quantum tunneling of magnetization.

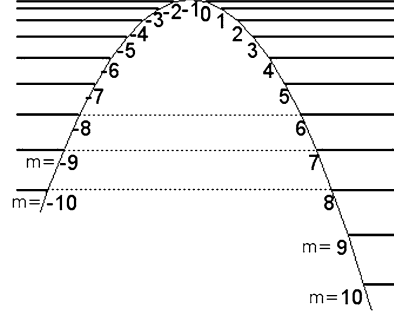
The spin Hamiltonian of the Mn-12 molecule subjected to the magnetic field, $\mathbf{B}$, in the direction of the easy magnetization axis z is

$$H = -DS_z^2 - g\mu_B S_z B_z + H_\perp \quad (1.2)$$

where D is the anisotropy constant, g is the gyromagnetic factor, μ_B is the Bohr magneton, and $H_\perp$ is a perturbation that does not commute with S_z . If $\mathbf{S}$ were a classical vector, the dependence of the classical energy of the molecule on the orientation of this vector, up to the small perturbation due to $H_\perp$, would be given by

$$E = -DS^2 \cos^2 \theta - g\mu_B S B \cos \theta \quad (1.3)$$

Fig. 1.5 Quantized spin energy levels of Mn-12 molecule at the second resonant field



where θ is the angle that $\mathbf{S}$ makes with the z -axis. The dependence of E on $\cos \theta$ is shown by the parabolic line in Fig. 1.5. It represents the magnetic anisotropy barrier discussed in the previous section. In quantum mechanics, however, the projection of $\mathbf{S}$ onto the z -axis is quantized, $S_z|m\rangle = m|m\rangle$, where m is an integer between -10 and 10 for $S = 10$. Consequently, the energy is quantized too,

$$E = -Dm^2 - g\mu_B Bm \quad (1.4)$$

The corresponding 21 quantized energy levels for $S = 10$ are shown by horizontal lines in Fig. 1.5.

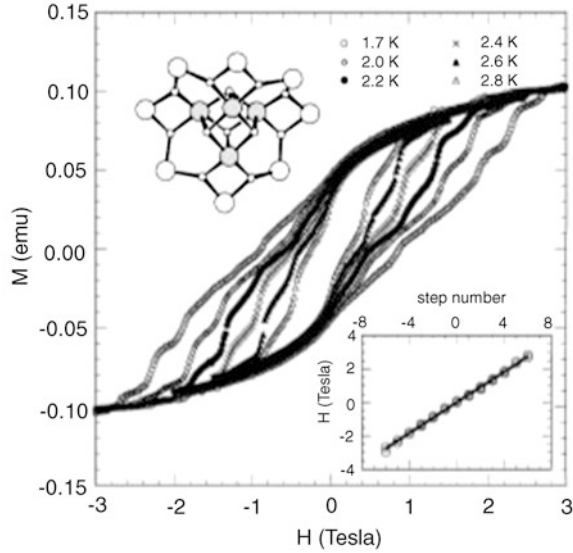
Imagine now that one magnetizes the crystal of Mn-12 molecules in the negative z -direction, making all molecules to occupy the level $m = -10$. If the field is now applied in the positive z -direction the $m = -10$ level becomes metastable, while $m = 10$ becomes the ground state, see Fig. 1.5. If no other interactions are involved, a transition, due to $H_\perp$ in (1.2), from a level on the left of the parabolic barrier to a level on the right of the barrier is possible only if the energies of the two levels coincide. Equating E of (1.4) for two levels, m and m' , one obtains that energies of pairs of m -levels coincide at discrete values of the magnetic field,

$$B_z = -(m + m')B_0, \quad B_0 = \frac{D}{g\mu_B} \quad (1.5)$$

Since m and m' are integers, it is easy to see that the corresponding resonances are equidistant on the magnetic field, separated by the field B_0 . For Mn-12 this field is about 0.45 T.

The above consideration makes it clear that at $T = 0$ the change of the projection of $\mathbf{S}$ on the easy magnetization axis may occur only via quantum tunneling between resonant m -levels at the discrete values of the magnetic field. Consequently the hysteresis curve of the Mn-12 acetate crystal will not be as smooth as the conventional hysteresis curve of magnetic materials because the bulk of the change of the magnetization should occur at the field that is multiple of B_0 . Finite temperature makes this effect less dramatic because of thermal overbarrier transitions. However, the steps in the magnetization at the values of the field that are multiple of B_0 should be apparent at a finite temperature as well because quantum tunneling adds to thermal

Fig. 1.6 This figure reports stepwise magnetization curves in Mn-12 acetate

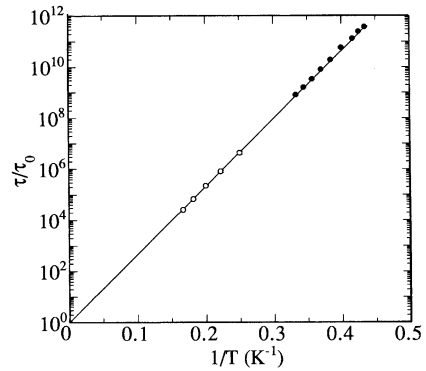


activation. (In Mn-12 thermal effects completely take over quantum transitions at temperature above 3 K.)

Early experiments on quantum tunneling of magnetization in Mn-12 performed in Grenoble tried to detect non-thermal magnetic relaxation at very low temperature. They were inconclusive because of the very low tunneling rate in Mn-12 at $T = 0$. Our first measurements of Mn-12 in New York and Barcelona were less ambitious. They were aimed at the accurate magnetic characterization of Mn-12 acetate through measurements of its magnetization curve. As it turned out this was all one needed to demonstrate unambiguously the existence of quantum spin tunneling in Mn-12. Quantum hysteresis curve of Mn-12, first observed by us [8] in dc magnetization measurements performed in New York in 1995, is shown in Fig. 1.6. Soon thereafter we performed in Spain the ac susceptibility measurements of Mn-12 [10] that confirmed the existence of resonant spin tunneling and permitted to extract the tunneling prefactor $\tau_0 \sim 10^{-7}$ s in the formula for the relaxation rate, $\tau = \tau_0 \exp[U/(k_B T)]$, see Fig. 1.7.

The importance of our first works on Mn-12 was not simply in the observation of equidistant magnetization steps but also in their correct interpretation as resonant spin tunneling along the lines of the theoretical argument presented above. That interpretation of the data was not straightforward at the time, it took us some time to sort things out. The problem was in the strong dependence of the height of the magnetization steps on temperature and on the rate at which the magnetic field was changed in experiment. Different steps kept appearing and disappearing on us depending on experimental conditions. Theoretical works appearing after our experiments suggested that this striking behaviour was based upon two main mechanisms: Dobrovitski and Zvezdin [25] noticed that the height of the magnetization step was determined by the Landau-Zener theory of adiabatic quantum transitions between

Fig. 1.7 Original figure [10] (1996) that permitted extraction of the tunneling attempt time from ac susceptibility measurements (*open circles*) and dc magnetic relaxation measurements (*closed circles*)

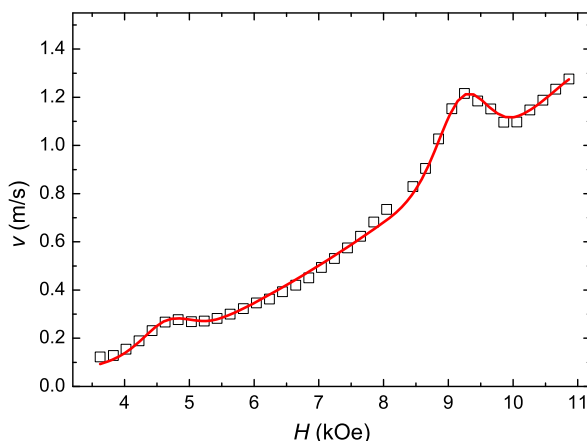


crossing energy levels, which explained the dependence on the field-sweep rate. On the other hand, the temperature dependence of the resonant tunneling effect was explained by means of the concept of thermally assisted spin tunneling, in which spins were first excited by phonons to higher energy levels and then tunnelled from these levels across the energy barrier [26–29].

For ten years after our discovery of resonant spin tunneling in Mn-12, one mystery about Mn-12 remained unsolved. In early 1990s Paulsen and Park, working in Grenoble, reported that sufficiently large crystals of Mn-12 exhibited abrupt reversal of the magnetization that did not seem to follow any clear pattern on temperature and magnetic field. This phenomenon received the name of “magnetic avalanche”. For quite a while it was considered an impediment to the measurements of quantum magnetization steps. In 2005 the group of Sarachik in New York placed micro-Hall sensors along the length of the Mn-12 crystal and observed that the avalanche propagated through the crystal as a narrow front moving at a constant speed [14]. Chudnovsky came up with an explanation in terms of the magnetic deflagration.

Deflagration is a technical term for slow combustion. A mixture of hydrogen and oxygen in a pipe would burn via a slow (compared to the speed of sound) propagation of a burning front inside which the chemical energy is released. In a Mn-12 crystal the role of the chemical energy is played by the Zeeman energy of the magnetic moment in the magnetic field. The rest is similar to the burning of a flammable substance. In Sarachik’s experiments deflagration was always triggered by the magnetic field sweep and occurred at random values of the field. This prevented Sarachik’s group from observing magnetic avalanches at resonant fields. In Barcelona we triggered avalanches by surface acoustic waves at a fixed value of the field. This allowed us to observe an interesting phenomenon that was not known for chemical combustion. We found that the “flammability” of the Mn-12 crystal increased at the resonant fields due to the quantum enhancement of the rate of the magnetization reversal [15], see Fig. 1.8. We called this phenomenon “quantum deflagration”.

Fig. 1.8 Quantum peaks in the field dependence of the velocity of the magnetic flame in a Mn-12 crystal [15]



1.4 Conclusion

As years went by, Mn-12 has been researched almost exhaustively, or has it been?

Chemists found that Mn-12 acetate molecules were not entirely identical as previously thought, but slightly different in $H_{\perp}$ due to solvent disorder. The exact form of $H_{\perp}$ was established in a series of precise measurements performed by the groups of Hill and Kent [30]. Another spin 10 molecular magnet, Fe-8, was discovered and soon thereafter chemists began producing new molecular magnets at an astonishing rate. They have been investigated theoretically by diagonalization of spin Hamiltonians [31, 32] and by density functional theory [33]. Topological interference in spin tunneling phenomena has been predicted [34] and observed [35]. Tunneling in antiferromagnetic and ferrimagnetic molecular clusters has been studied [36–38]. Effects of nuclear spins and dipolar interactions in molecular magnets were addressed [39–43] and magnetocaloric effects have been investigated [44, 45]. Crossover between quantum and thermal regimes has been investigated in some detail [46–50]. Resonant interactions of molecular magnets with electromagnetic radiation has been measured [51–59]. Also long-range dipolar ordering has been studied [60–65]. Behavior of Mn-12 in ultrafast pulses of the magnetic field has been investigated [66, 67]. Magnetic molecules on surfaces have been studied [68] and conduction through single molecules placed between conducting leads has been measured. This account is, to the best of my knowledge, accurate. As the field of quantum magnetic relaxation and spin resonant tunneling goes back to the end of the 1980s, there will doubtless be some recollections that I have failed to fully acknowledge. For these mistakes, I sincerely apologize in advance.

Are any important discoveries left in this field for the younger generation of researchers? One area that remains cloudy is the possibility of superradiant emission of electromagnetic radiation by a crystal of molecular magnets. While we and others have published papers in this field [69, 70] the definitive proof of this possibility (or impossibility) is still absent. The distances between the spin levels of many molecular magnets are in the sub Terahertz range which has multiple applications. It is

not clear, however, whether random dipolar and nuclear fields would allow strong coherent effects. Experiments inside a resonant cavity may help to figure this out. Up to date the reversal of the magnetic moment in molecular magnets has been explained claiming it occurs at the level of individual molecules [71]. However, it may be the case that this reversal process could occur at the level of clusters of molecules induced by spin-phonon transitions.

Since the energy barrier between opposite orientations of the magnetic moment is formed by weak relativistic interactions, a crucial question would be whether stable molecular magnets can ever break liquid nitrogen temperature of 77 K. Molecules with big magnetic moments, such as those containing rare earth atoms, may have their magnetization frozen at 77 K because of the strong magnetic anisotropy. Making identical molecules comparable to mesoscopic magnetic particles will be a challenging task for chemists. Another challenging question would be whether magnetic molecules can ever become ultimate memory units of conventional computers or even elements of quantum computers. I hope to see answers to these questions in the near future.

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