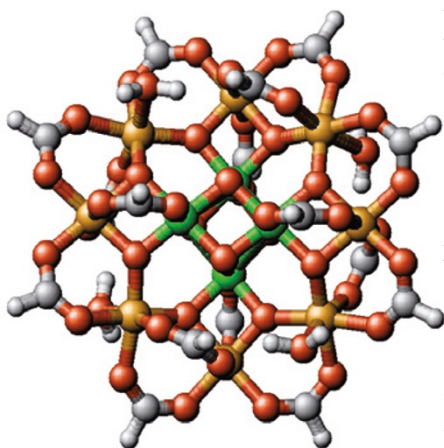


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MILESTONE 22

The difficult middle ground



The molecular magnet Mn_{12} . Image courtesy of Jens Kortus, Tunna Baruah and Mark Pederson.

Much of physics lends itself to a convenient division between big and small: the macroscopic classical world and the microscopic world of the quantum. Yet the transition between these spheres of influence is far from clear-cut, and increasingly we are seeing examples of phenomena that are manifestly quantum mechanical encroaching into territory once believed to be the exclusive purview of classical physics. Spin is no exception.

The year 1996 saw the publication, by Jonathan Friedman *et al.* and Luc Thomas *et al.*, of what many consider to be the definitive proof that mesoscopic spin systems (that is, systems with sizes or numbers of component spins that place them firmly in the conceptually difficult middle ground) can undergo quantum-mechanical tunnelling of their total magnetization. These experiments revealed transitions between bulk magnetic states that were not driven by thermal fluctuations, as one would expect classically; instead,

they corresponded unambiguously to quantum-mechanical tunnelling events between different collective spin states of the whole system that have been brought into resonance by an applied magnetic field.

The idea that macroscopic (or rather mesoscopic) quantum phenomena might be realized in magnetic spin systems has a long and convoluted history, with hints of such behaviour stretching back at least to work by Bernard Barbara and colleagues in 1972. However, it was not until the 1980s that such ideas were placed on a firm theoretical footing. Yet more experimental results suggestive of mesoscopic quantum behaviour were soon forthcoming; however, the response of the physics community remained largely ambivalent, even sceptical, about the central claims.

A key problem with the early experiments was that any tunnelling effects in the thin films and quantum dots utilized in these studies could be readily masked by other factors — such as a distribution of energy barriers between magnetic states — rendering any ‘proof’ of mesoscopic quantum behaviour ambiguous. What was needed was an experimentally ‘clean’ system: either the ability to probe experimentally the magnetization properties of a single mesoscopic particle or the availability of an ensemble of strictly identical particles.

In the meantime, chemist Roberta Sessoli and colleagues were investigating small molecular clusters containing several manganese ions that were magnetically coupled to give a high-spin ($S = 10$) ground state. The motivation for their 1993 work was to develop, using the powerful ‘bottom-up’ methods of chemical

synthesis, well-defined molecular materials with potentially useful magnetic properties. Yet these chemists also had in their hands, or rather in their test-tubes, the very thing that the physicists needed to realize convincingly mesoscopic quantum effects: ensembles of identical magnetic molecules.

By 1995, Miguel Novak and Sessoli, and Barbara and colleagues, had the first evidence of resonances between the high-spin states of these molecular magnets; and, barely a year later, came the smoking-gun demonstration of mesoscopic quantum tunnelling. Now the pursuit of exciting new physics could begin...

*Karl Ziemelis, Chief Editor
Physical Sciences, Nature*

ORIGINAL RESEARCH PAPERS Barbara, B., Fillion, G., Gignoux, D. & Lemaire, R. Magnetic aftereffect associated with narrow domain walls in some rare earth based intermetallic compounds. *Solid State Commun.* **10**, 1149–1151 (1972) | Enz, M. & Schilling, R. Spin tunneling in the semiclassical limit. *J. Phys. C* **19**, 1765–1770 (1986) | van Hemmen, J. L. & Suto, A. Tunneling of quantum spins. *Europhys. Lett.* **1**, 481–490 (1986) | Chudnovsky, E. M. & Gunther, L. Quantum tunneling of magnetization in small ferromagnetic particles. *Phys. Rev. Lett.* **60**, 661–664 (1988) | Sessoli, R., Gatteschi, D., Caneschi, A. & Novak, M. A. Magnetic bistability in a metal-ion cluster. *Nature* **365**, 141–143 (1993) | Barbara, B. *et al.* Mesoscopic quantum tunneling of the magnetization. *J. Magn. Magn. Mater.* **140**, 1825–1828 (1995) | Novak, M. A. & Sessoli, R. in *Quantum Tunneling of Magnetization — QTM '94*. NATO ASI Series E: Applied Sciences Vol. 301 (eds Gunther, L. & Barbara, B.) 171–188 (Kluwer Academic, Dordrecht, 1995) | Friedman, J. R., Sarachik, M. P., Tejada, J. & Ziolo, R. Macroscopic measurement of resonant magnetization tunneling in high-spin molecules. *Phys. Rev. Lett.* **76**, 3830–3833 (1996) | Thomas, L. *et al.* Macroscopic quantum tunnelling of magnetization in a single crystal of nanomagnets. *Nature* **383**, 145–147 (1996)

FURTHER READING Stamp, P. C. E., Chudnovsky, E. M. & Barbara, B. Quantum tunneling of magnetization in solids. *Int. J. Mod. Phys. B* **6**, 1355–1473 (1992) | Barbara, B. & Gunther, L. Magnets, molecules and quantum mechanics. *Phys. World.* **12** (3), 35–39 (1999)