Neutron scattering from quantum condensed matter

Steven T. Bramwell and Bernhard Keimer

Collective quantum phenomena such as magnetism, superfluidity and superconductivity have been pre-eminent themes of condensed-matter physics in the past century. Neutron scattering has provided unique insights into the microscopic origin of these phenomena.

Neutrons are impervious to the Coulomb interaction, so penetrate deeply into most materials. There, they sense the position of atomic nuclei through the strong interaction and magnetic moments through the dipole–dipole interaction. The scattering cross-sections in both channels are precisely understood, so theorists need not be concerned with modelling or simulating the scattering process. Instead, they can focus on computing intrinsic density and spin-correlation functions, which can be translated into the neutron cross-section by means of simple analytical expressions. Because neutrons produced by high-flux sources have wavelengths comparable to interatomic distances and energies comparable to typical collective excitations in condensed matter, both static and dynamic correlations are probed precisely in the spatial and temporal range of greatest interest. Exchange and collaboration between neutron scatterers and condensed-matter theorists has thus been an exceptionally fertile ground for cultivating new science.

This story began with Clifford Shull's introduction of elastic neutron scattering as a way to directly visualize the microscopic arrangement of spins on the atomic scale[1]. Such information is indispensable for any model of magnetism in solids, and advances in instrumentation over the past half-century have allowed the determination of increasingly complex magnetic structures — including, for instance, mesoscopic textures of topological defects such as vortices[2] and skyrmions[3]. These experiments can be performed under a large variety of experimental conditions, including temperatures that span more than 12 orders of magnitude — from below 1 nK, to above 1,000 K, as illustrated by studies of nuclear antiferromagnetism in silver[4] and electronic ferromagnetism in iron[5], respectively. Inelastic neutron scattering now provides detailed maps of dynamical correlations in fluctuating quantum systems such as low-dimensional magnets, superfluids and superconductors. Many of these experiments were motivated by theoretical predictions; in turn, they have driven advances in theoretical research.

Here we present a personal selection of neutron scattering experiments that have inspired our own work. Without any claim of completeness, they highlight some of the unique contributions of neutron scattering to the development of condensed-matter science.

Model magnets

To understand the physics of a magnetic material is like crossing a torrent on a series of stepping stones. On the near bank one has microscopic physics and on the far bank, macroscopic experiment. A few sure steps brings one to a plausible 'spin Hamiltonian', but then there is a particularly disconcerting leap to be made to use this Hamiltonian to calculate measurable properties. The scale of the leap is brought home when one realizes that spin Hamiltonians generally start with an apparently innocuous request (denoted by the symbol $\Sigma_{i,j}$) to sum over all ~10^{23} spins in a typical sample.

The mean-field theory of Weiss, Néel and Landau replaces all but a handful of these 10^{23} spins by their mutual average, and thereby guarantees a safe passage to the far bank. It is certainly a successful theory, predicting a transition to magnetic order below a critical temperature, as observed in thousands of magnetic

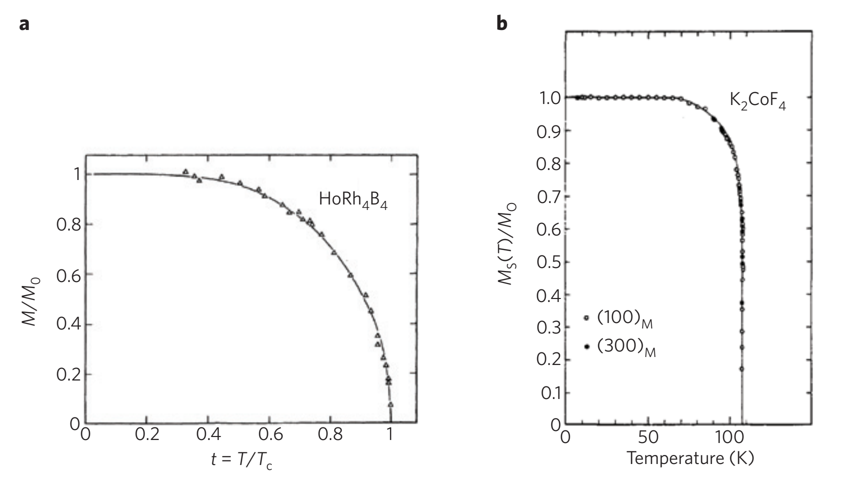

Figure 1 | Canonical model magnets. **a**, A magnet that ideally obeys mean-field theory in several thermodynamic quantities[6]. Here the square root of the neutron magnetic Bragg scattering of $HoRh_4B_4$ (M) is compared to the mean-field prediction $M = M_0\,(T_c - T)^{1/2}$, where T_c is the critical temperature. **b**, The equivalent quantity for the layered magnet K_2CoF_4 (ref. 7). The line corresponds to Onsager's famous exact solution of the two-dimensional Ising model (as first published by Yang)[9]. Figure reprinted with permission from: **a**, ref. 6, © The American Physical Society; **b**, ref. 7, © Elsevier.

materials. However, it does not encompass all magnetic behaviour, and if relied on too much, can deceive one as to the true scope of magnetism. Alternative strategies focus on actually solving the many-body problem posed by the troublesome summation. It is not possible to achieve this for a general spin Hamiltonian, but it can be achieved (usually with great effort) for simplified, idealized ones.

Real magnetic materials that approximate these solvable Hamiltonians serve as archetypes of our understanding of magnetism. We know about these 'model magnets' primarily because neutron scattering has opened up the gigantic chemical parameter space of magnetic materials to detailed experimental scrutiny. Most magnetic materials have antiferromagnetic interactions, which make them obscure to many experimental probes. However, neutron scattering is tailor-made for studying such systems, and has been used to image their magnetic order, correlations and excitations in great detail. By identifying model magnets and studying them with neutron scattering, we have been able to break away from the mean-field paradigm, and to reveal the full richness of magnetic behaviour — a process that is ongoing, and far from complete.

Figure 1 shows two striking comparisons between theory and experiment. In both cases the quantity measured by neutron scattering is a magnetic 'order parameter' — magnetization in the case of ferromagnetic $HoRh_4B_4$ (ref. 6) and staggered magnetization of antiferromagnetic K_2CoF_4 (ref. 7). For $HoRh_4B_4$, the line is the prediction of mean-field theory. Its detailed success — which is the exception rather than the rule for magnetic materials — can be associated with long-range spin interactions that make mean-field theory nearly exact in this instance. For K_2CoF_4, the line corresponds to Onsager's famous exact solution of the two-dimensional (2D) Ising model, the prototypical magnetic model with short-range interactions[8,9]. K_2CoF_4 (ref. 7) is a layered material with strong interactions within each layer and much weaker interactions between the layers, hence the 2D Ising model applies.

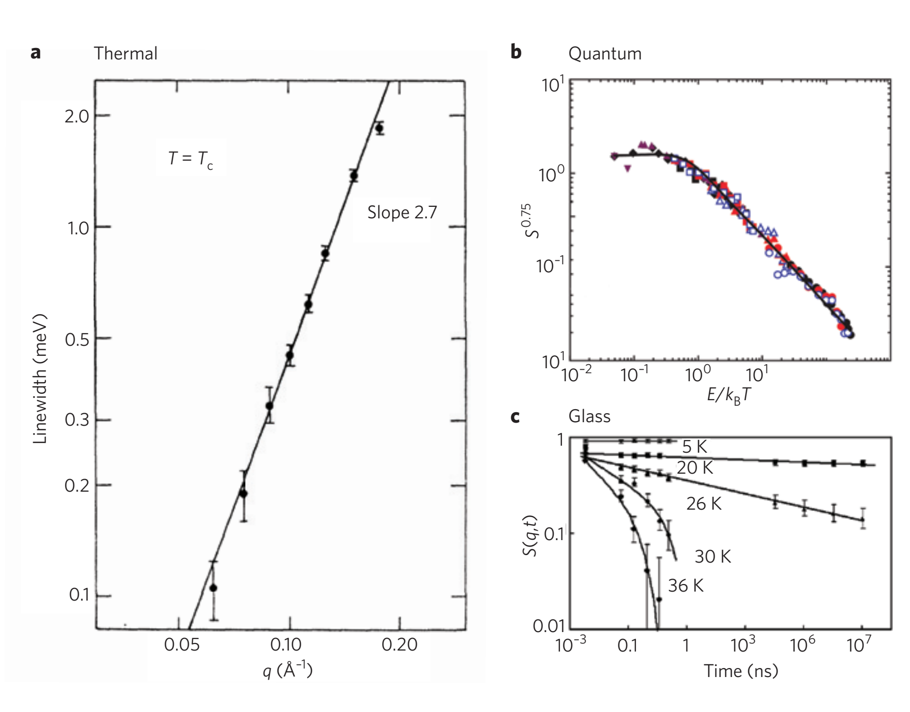

Figure 2 | Dynamical scaling revealed by inelastic neutron scattering. **a**, Universal energy momentum relation of critical fluctuations in a ferromagnet, illustrated by experimental results on iron at its Curie temperature[5]. **b**, E–T scaling of the scattering function $S(q, E)$ at the quantum critical point of the heavy-fermion antiferromagnet $CeCu_{6-x}Au_x$ with $x = 0.1$ (ref. 14). The data points shown were measured at different excitation momenta. **c**, Neutron spin-echo and a.c. susceptibility data showing power-law time dependence of the dynamical susceptibility (proportional to $S(q, t)$) of the canonical spin glass $Cu_{0.95}Mn_{0.05}$ at its transition temperature, 26 K (ref. 11). S, scattering function; E, energy; q, momentum; t, time. Figure reprinted with permission from: **a**, ref. 5, © The American Physical Society; **b**, ref. 14, Nature Publishing Group; **c**, ref. 11, © The American Physical Society.

Near a critical point, where the order parameter goes to zero, spatial- and time-dependent correlations obey universal power laws and scaling relations. In fact, these properties characterize not only thermal critical points, as shown in Fig. 1, but also, for example, glassy behaviour and phase transitions driven by quantum fluctuations. Inelastic neutron scattering affords a unique probe of distance- and time- (or equivalently energy and momentum) dependent scaling laws in such systems. Figure 2 shows three examples: the energy–momentum relation for spin fluctuations in iron at its critical point, the power-law decay of correlations over nine decades in time for a spin glass, and the scaling of the energy (E)-dependent susceptibility with E/T (where T is temperature) near a quantum critical point — a striking universality that has been observed at quantum critical points in a wide variety of materials including insulators[13], metals[14] and superconductors[15].

The existence of quantum critical points implies that there are systems with quantum fluctuations strong enough to suppress long-range order even when approaching zero temperature. In 1931, Bethe showed theoretically that a simple chain of spins $S = 1/2$ interacting with antiferromagnetic Heisenberg exchange typifies this behaviour[16]. Many magnetic materials with chain-like networks of spins indeed show greatly suppressed magnetic ordering temperatures, with the usual ordered state being replaced by an extended temperature range of nearly ideal 1D behaviour. Neutron scattering experiments on these materials can elucidate the nature of excitations out of such a quantum-disordered ground state.

The ordered states arising from the mean-field paradigm naturally support wave-like excitations such as lattice vibrations in a crystal and spin waves in a magnet. Quanta of these excitations are phonons with $S = 0$ and magnons with $S = 1$. As quasiparticles with even spin they are therefore classified as bosons rather than fermions.

For many years theoreticians naturally assumed that the excitations in 1D Heisenberg chains would likewise be spin waves, despite noting a strong dependence on the spin-wave spectrum on the spin value[17,18]. Neutron scattering experiments on $(CD_3)_4NMnCl_3$, with $S = 5/2$, confirmed that the large spin of Mn^{2+} in this material led to a classical spin-wave spectrum[19], but in $CuCl_2{\cdot}2NC_5D_5$ with the smaller effective spin of Cu^{2+}, $S = 1/2$, an unusual line shape hinting at an unexpected continuum of excitations was reported[20,21]. Then in 1981, Fadeev and Takhtajan[22] realized that for the $S = 1/2$ chain, a conventional magnon with spin $S = 1$ should split apart or 'fractionalize' into two fermions with spin $S = 1/2$. Excitations from the quantum-disordered ground state were suggested to be fermions, not bosons, as in the standard picture.

This prediction was confirmed by neutron scattering[23]. The fermonic quasiparticles, now known as spinons, are thermally excited in pairs and are the quantum equivalent of classical domain walls or solitons. Their direct excitation by neutrons gives rise to the continuum of scattering noted in the early experiments. A recent and beautiful measurement of the spinon continuum for a model 1D Heisenberg chain[24] is shown in Fig. 3a, where another striking match between theory and experiment is observed; this data also confirmed the existence of higher-order spinon states. Perhaps ironically, this exotic many-body quantum behaviour is observed in what most would consider a far from exotic material, namely, deuterated copper sulphate, $Cu(SO_4){\cdot}5D_2O$.

Onsager's solution of the 2D Ising model and Bethe's solution of the 1D Heisenberg chain form perhaps two of the three cornerstones of exactly solved models of statistical mechanics. The third class is classical ice-type or vertex models, in which exact solutions, for example by Lieb[26] and Baxter[27] illustrate further criteria for disordered low-temperature states and unusual critical behaviour. The discovery of 'spin ice' by neutron scattering[28] showed that ideal model magnets in this class can be realized experimentally. Spin ice, like the 1D quantum antiferromagnet, has a disordered low-temperature state and fractionalized quasiparticles, yet it achieves this in a dense 3D spin structure with essentially ferromagnetic interactions[29]. In spin ice, the quasiparticles behave like fractionalized dipoles, or magnetic monopoles[30,31]. Evidence for such excitations was indeed extracted from neutron-intensity maps obtained from spectrometers equipped with modern multidetectors[32–34], and a detailed comparison with theoretical predictions (Fig. 3b).

Present interest focuses on the possibility of discovering a 'quantum spin ice' — a spin ice system with strong quantum fluctuations[25]. In addition to a band of monopoles — longitudinal

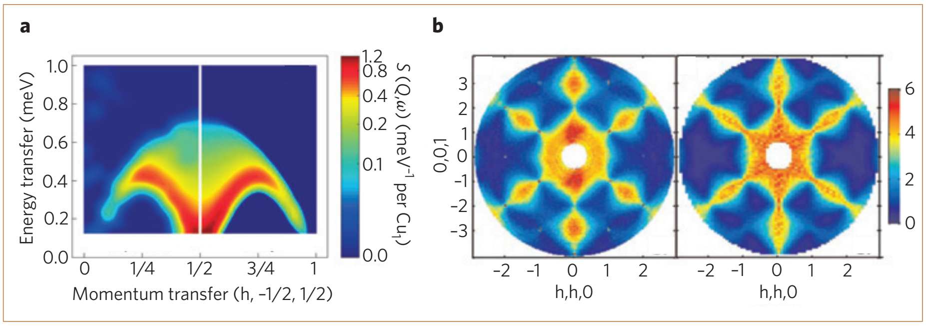

Figure 3 | Fractionalized excitations in model magnets. **a**, Spinon continuum measured by neutron scattering in copper sulphate. Left, experiment; right, theory[24]. $S(Q,\omega)$ is the dynamic structure factor. **b**, Diffuse neutron scattering maps of magnetic intensity in the spin-ice material $Ho_2Ti_2O_7$. Left, experiment; right, theory[32]. The width of the narrow 'pinch point' features measure the density of emergent monopole excitations. Figure reprinted with permission from: **a**, ref. 24, Nature Publishing Group; **b**, ref. 32, © American Association for the Advancement of Science.

excitations — there is also a linearly dispersed band of transverse, wave-like excitations, behaving like photons. Despite some promising candidates, most notably $Pr_2Zr_2O_7$ (ref. 35), such a spectrum has not yet been clearly observed in experiment, but doing so would add an exciting new dimension to model magnetism[36].

Superfluidity and superconductivity

Entirely novel forms of order can emerge out of the quantum-disordered ground state of a many-particle system. A classic example is superfluidity, which emerges out of the liquid state of an ensemble of ^4He atoms. Quantum zero-point motion of these light, electronically inert, bosonic atoms obliterates crystalline order even in the limit of zero temperature, in a manner analogous to spin fluctuations in 1D model magnets. To explain the unique macroscopic properties of helium below the 'lambda transition' at $T = 2.2$ K, Landau proposed two sets of bosonic quasiparticles: phonons with dispersion that are linear in momentum (q) and rotons with a dispersion minimum at $q \neq 0$. Pioneering inelastic-neutron-scattering experiments confirmed this prediction[37] (Fig. 4), and thus an exceptionally fruitful dialogue was started between experimentalists and theorists on this canonical quantum many-body system.

In this regard, the direct observation of the macroscopic population of the $q = 0$ wavefunction in the superfluid was an important milestone[38]. This key signature of Bose condensation appears in the momentum distribution $n(q)$, which can be obtained from the phonon–roton spectra by energy-integration. The $n(q)$ plot shown in Fig. 4b is the analogue of the images of expanding clouds of Bose-condensed ultracold atoms that gained such notoriety in the 1990s. The neutron data directly determine the superfluid condensate fraction (that is, the fraction of atoms in the $q = 0$ state), which is reduced to only about 8% as a consequence of interactions between the helium atoms[39]. Recent experimental achievements made possible by advanced instrumentation include the accurate measurement of roton lifetimes[42], the observation of rotons in monolayers of fermionic ^3He atoms[43], and the exploration of model magnets as analogues of Bose condensation[13].

Neutron scattering has also served as one of the most influential probes of superconductivity, an equally fascinating macroscopic quantum phenomenon. According to the Bardeen–Cooper–Schrieffer theory, the superconducting state is a condensate of electronic 'Cooper pairs' bound together by an intermediate boson. The isotope effect (that is, the dependence of the superconducting transition temperature on the nuclear mass) led Bardeen, Cooper and Schrieffer to identify phonons as the 'pairing glue', but conclusive confirmation of this hypothesis had to await careful comparison of anomalies in electronic tunnelling spectra[44] and the phonon spectrum obtained by neutron scattering[45]. In an elegant set of neutron scattering experiments on the conventional superconductor Nb_3Sn, Axe and Shirane[46] demonstrated the converse effect, namely the renormalization of the phonon energies and linewidths in the superconducting state (Fig. 4c). This effect arises because the electron–phonon interaction, which broadens the phonon profiles in the normal state, becomes inactive for phonons with energies lower than the superconducting energy gap. The q-dependent phonon linewidths thus provide a direct signature

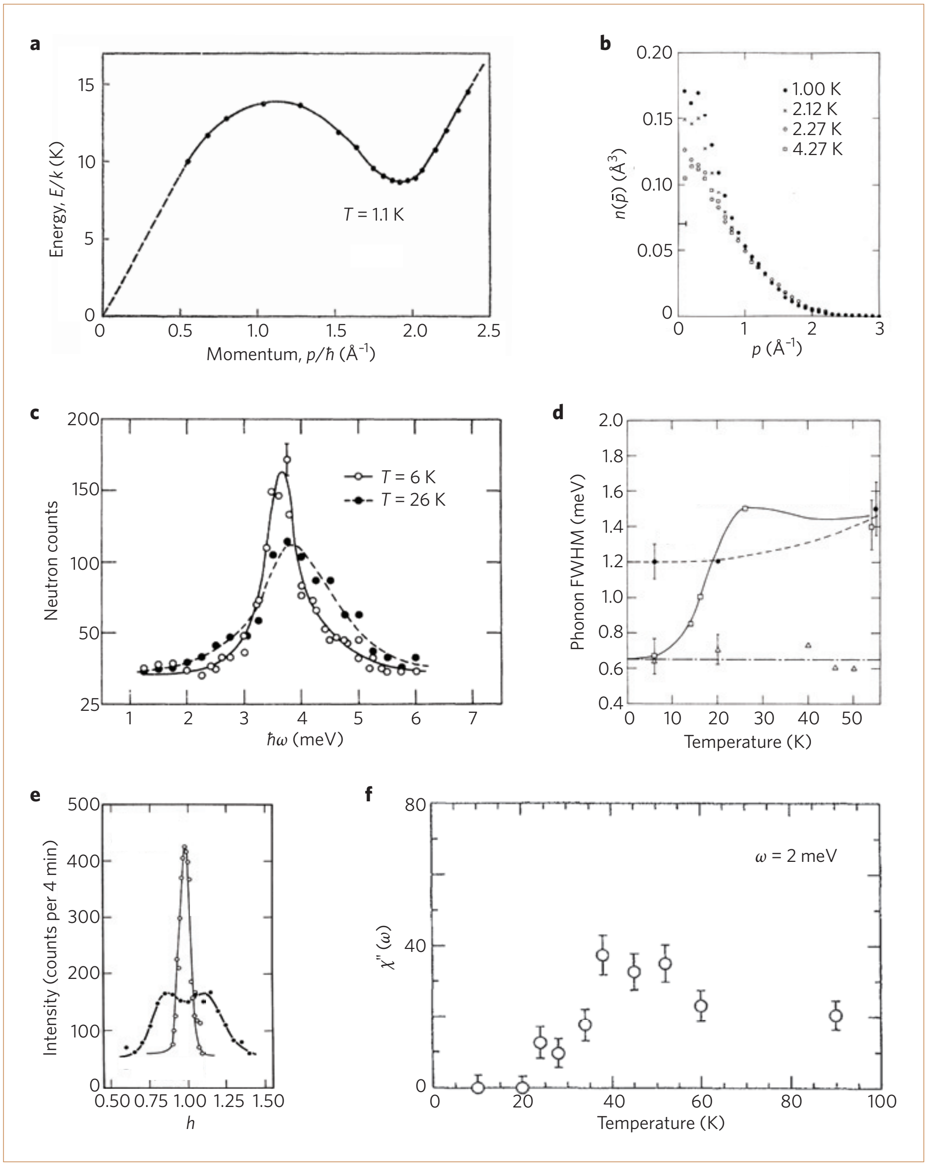

Figure 4 | Neutron scattering from superfluids and superconductors. **a**, Dispersion relation of phonons and rotons in liquid helium extracted from early inelastic neutron scattering experiments[37]. **b**, Momentum distribution of helium in its normal liquid and superfluid states[38]. **c**,**d**, Phonon profiles (**c**) and q-dependent phonon linewidths above and below T_c in the conventional superconductor Nb_3Sn (**d**, ref. 46). The curves in **d** are linewidths of different phonon modes. **e**, Spin fluctuation profiles in the unconventional superconductor $La_{1.85}Sr_{0.15}CuO_4$ (filled circles) and the antiferromagnetic parent compound La_2CuO_4 (open circles; ref. 49). **f**, Temperature dependence of the dynamical spin susceptibility χ'' (proportional to the scattering intensity) at excitation energy 2 meV in $La_{1.85}Sr_{0.15}CuO_4$ with superconducting $T_c = 37.3$ K (ref. 50). FWHM, full-width at half maximum. Figure reprinted with permission from: **a**, ref. 37; **b**, ref. 38; **c**,**d**, ref. 46; **e**, ref. 49; **f**, ref. 50, © by The American Physical Society.

of the superconducting gap (Fig. 4d). Follow-up experiments with μeV energy resolution now allow quantitative tests of *ab initio* calculations of the electron–phonon interaction in metals[47].

The power of neutron scattering as a probe of 'unconventional' superconductors became obvious immediately after the discovery of high-temperature superconductivity in 1986. Magnetic neutron-scattering experiments first demonstrated that the undoped parent compounds of the copper oxide superconductors are antiferromagnetic insulators with conventional spin-wave excitations[48]. Soon after, they showed that incommensurate spin excitations with intensity comparable to spin waves persist as these materials are doped into the metallic regime, even though magnetic long-range order is no longer present (Fig. 4e)[49]. This discovery has inspired a slew of models that describe the 'normal' state of the superconducting cuprates as a quantum-disordered spin fluid, a state that is fundamentally distinct from an ordinary metal.

Another milestone was reached when neutron experiments revealed a massive rearrangement of the magnetic spectrum below the critical temperature T_c (ref. 50), one that is much stronger than the subtle superconductivity-induced phonon renormalization in conventional superconductors (Fig. 4d). This discovery highlighted spin fluctuations as a key player in the mechanism of high-T_c superconductivity. Figure 4f shows that the magnetic spectral weight in superconducting $La_{1.85}Sr_{0.15}CuO_4$ actually disappears entirely below the superconducting energy gap. Experiments on other cuprates[51] later showed that the low-energy spectral weight accumulates in high-energy 'resonant modes' akin to the $q = 0$ mode in superfluid helium (Fig. 4b), which are hallmarks of the d-wave symmetry of the superconducting gap function. In contrast to phonons in conventional superconductors, these magnetic modes are generated by the strongly correlated electrons themselves. Together with closely similar effects observed in iron pnictides and heavy-fermion compounds[52], magnetic modes serve as key experimental cues in the ongoing quest for a theory of unconventional, electronically driven superconductivity.

Outlook

The above examples are drawn from the vast literature that documents the extraordinary power of neutron scattering as a probe of quantum condensed matter. Research in this field continues to advance rapidly on many fronts, including materials discovery, experimental methodology, and analytical and numerical computation. With brighter neutron sources[53], innovative techniques such as coherent manipulation of the neutron spin[11,47] and advanced sample environments such as high-field magnets, neutron scattering will continue to play a commanding role in this ongoing research adventure. ❐

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Reinventing neutron science in Europe

Dimitri N. Argyriou

Neutron science has been a remarkable success story for European research. For this to continue, scientists need to be prepared to forge new networks and technologies.

Recently, after a difficult meeting and over a bottle of red wine, a colleague sighed and said (paraphrasing a little), "…in my time we turned the reactor on in the morning and off in the evening. Experiments took as long as needed. We had time to think. Now everything has to be done fast, fast, fast!" What my friend was reminiscing about is what many saw as the golden age of neutron science for studying novel states of matter or fundamental physics. This was a time when many countries were furnished with small- to medium-flux nuclear reactors bred out of the promises of the nuclear age, and scientists operated in a relaxed environment with the resources to experiment and innovate.

However, an equally important and often overlooked consequence of the presence of small national and regional neutron sources scattered around the continent is that they helped to form a strong and diverse community of scientists that debated, supported and built some of the world's most successful large-scale facilities, such as the Institute Laue Langevin in Grenoble, France, and ISIS in Harwell in the UK, and more recently the Meir-Leibnitz Center near Munich, Germany. This led to a two-tier structure, with smaller neutron sources often tightly coupled to universities feeding experiments, ideas, people and innovation to the larger and more capable internationally oriented facilities. Although initially unintended, this state of affairs was nurtured, and it evolved in such a way that Europe eventually played host to the most sophisticated and advanced environment for science with neutrons in the world.

Dark clouds

Worryingly, shifting national budgets, ageing reactors and the events in Fukushima are putting unyielding pressure on this unique and successful model, hampering ideas and innovation. Some medium-flux reactors have already closed, while others such as the successful BER-II reactor in Berlin, Germany will close in 2020. Unfortunately, there is also speculation and uncertainty over the fate of other medium-sized neutron facilities. Reactors are not getting any younger and the general public is as suspicious of nuclear technology as it has ever been.

As the *Nature Milestones in Crystallography*[1] and the other Commentaries[2–4] in this issue testify, it is clear that neutrons have been central for understanding matter on both the microscopic and the macroscopic level. Although this is being recognized in Europe — as demonstrated by the investment in what will be the world's brightest source of neutrons, the European Spallation Source (ESS) in Lund, Sweden — careful thought is needed to ensure that the neutron science community remains able to experiment, innovate and grow.

The ESS will be a breakthrough neutron facility in many ways. Its accelerator will deliver a higher proton current than anything ever built for this purpose and this means more neutrons. The moderator package of the ESS, which is undergoing