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Progress in Physics (32)

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Steve Johnson [4], Rafael Abela [5], Christian Rüegg [1,6]

[1] Laboratory for Neutron Scattering, Paul Scherrer Institute, Villigen PSI
[2] Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institute, Villigen PSI
[3] Laboratory for Synchrotron Radiation, Paul Scherrer Institute, Villigen PSI
[4] Institute for Quantum Electronics, ETH Zürich
[5] SwissFEL, Paul Scherrer Institute, Villigen PSI
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Particles like neutrons, muons, and photons are excellent probes for studies of the complex structural, electronic and magnetic properties of matter. The Swiss Spallation Neutron Source (SINQ), the muon source and the Swiss Light Source (SLS) at the Paul Scherrer Institute provide intense beams of neutrons, muons and photons for condensed matter research. New instrumentation at these facilities and the free-electron laser SwissFEL, whose construction now started, offer unequaled opportunities for studies of the properties of materials at all length, energy and time scales.

New neutron spectrometer EIGER

Neutron spectroscopy with its cross section that is known exactly is an exceptional technique for studies of the elementary excitations of condensed matter systems. Up to now there were four spectrometers using cold neutrons from SINQ, but none for neutrons from its thermal moderator. The cold spectrometers are two triple-axis spectrometers (RITA-2 and TASP), one direct geometry time-of-flight spectrometer (FOCUS) and one inverse geometry time-of-flight backscattering instrument (MARS). Cold spectrometers are best suited to study excitations up to energies of about 5 meV. However, there is also a strong demand for experiments, which require larger energy transfers. In 2012 construction of the new thermal triple-axis spectrometer EIGER was completed. EIGER will extend the available energy range up to 50 meV.

The most important quality factors for a triple-axis-instrument are intensity and (low) background. Since the new instrument is the first thermal triple-axis instrument at a continuous spallation source it was a special challenge to design proper shielding for the monochromator in order to keep the fast neutron and gamma-ray background sufficiently low. Additionally, it is very desirable to be able to perform experiments with strong cryo-magnets, therefore, the shielding had to be built from non-magnetic materials. The final design is an optimized composite structure of different materials: boron for the absorption of thermal neutrons, tungsten for the absorption of fast neutrons and the attenuation of gamma-rays and additional special heavy (non-magnetic) concrete to absorb gamma radiation.
The principle layout for the primary spectrometer controlling the incident neutron beam consists of a sapphire filter, which reduces the contribution of fast neutrons at energies above 80 meV, an adjustable virtual source with a maximum width of 40 mm and a double focusing monochromator. The double focusing monochromator is made of 15×9 pieces of pyrolytic graphite (PG) with a mosaicity of about 30' and a size of 20×20 mm^2 each. The graphite is fixed on Al-lamellas, which can be rotated for horizontal focusing of the beam and bent for vertical focusing. As the secondary spectrometer, analyzing the energies of the scattered neutrons also by Bragg scattering from PG crystals, components of the former cold triple-axis spectrometer Drüchal at SINQ are used. A picture of the instrument is presented in Figure 1, with a helium cryostat (orange) at the position of the sample.

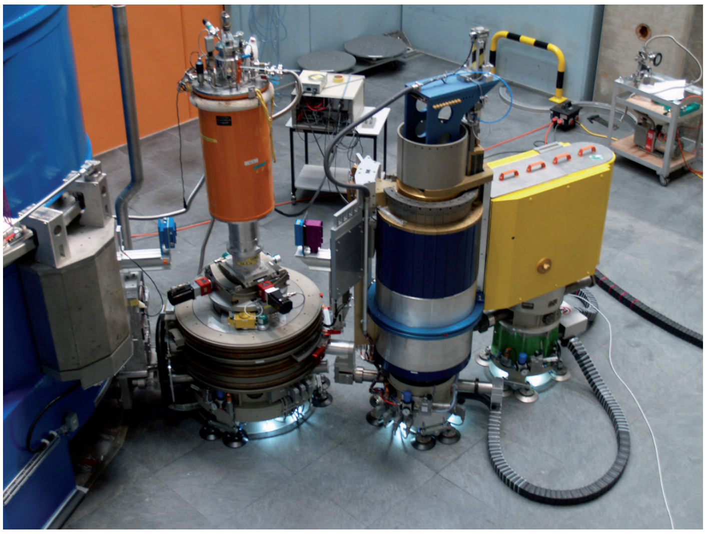

Figure 1: The new thermal triple-axis spectrometer EIGER at SINQ.

The resolution of the instrument at a final energy of 14.7 meV with double focusing monochromator and horizontal focusing analyzer was determined to 0.64 meV, in agreement with Monte-Carlo simulations of the instrument, and the total flux at the sample position was estimated with a calibrated monitor ($5.8 \cdot 10^6$ n·cm^{-2}·sec^{-1} at the same final energy).

During the commissioning phase in 2012 half a dozen experiments have already been performed on EIGER. When normalized by the source flux, its performance is among those of the best spectrometers of this type in the world. Efficiency gains for experiments at SINQ in the energy range 2-10 meV will be significant and new science will be done on materials with excitations at energies above that range and up to 50 meV, which were previously inaccessible. Among the first experiments were studies of magnetic excitations and phonons in quantum and frustrated magnets, magnetic semiconductors and novel superconductors. The unique combination of neutron and photon spectroscopy (like RIXS described below) at PSI and their complementarity now enable studies of the dynamics in solids from 0 to several eV on samples that are as diverse as single atomic layers of model two-dimensional insulators and large single crystals of superconductors.

New high-field µSR instrument

Spectroscopy with muons is another highly sensitive technique available at PSI. A worldwide unique, new instrument providing magnetic fields up to 9.5 T (with 0.1 mT homogeneity) and low temperatures (down to 20 mK) has been developed and installed at a dedicated beam line, as shown in Figure 2. This instrument makes use of a ~28 MeV/c muon beam. For most experiments, the spin of the originally fully longitudinally polarized beam is rotated by 90° with two "spin-rotators". Detecting the muon decay and measuring a spin frequency in ~10 T field represented a stringent requirement for the µSR spectrometer. The small curvature radius of the decay positron (10-20 mm) and the high Larmor precession frequency (1.3 GHz) required the development of a very compact spectrometer using detectors with very high time resolution that work in a high-field environment. The spectrometer developed at PSI has a diameter as small as 32 mm and an overall time resolution of 80 ps, provided by avalanche photo diode detectors. This is about an order of magnitude better than what is achieved in conventional spectrometers. Another challenge is the wish to insert in the horizontal bore of the magnet a dilution refrigerator. In collaboration with a company a pioneering solution was found leading to the development of a purely horizontal dilution refrigerator. The new high-field µSR instrument will begin user operation during 2013.

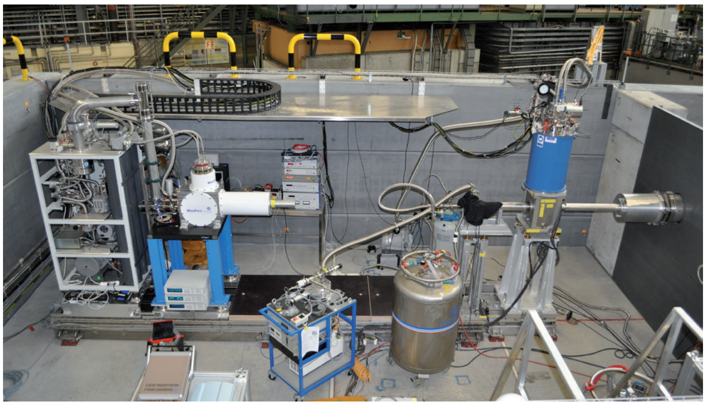

Figure 2: The new instrument for high-field µSR at PSI. The 9.5 T magnet is on the right (blue). On the left the dilution refrigerator with gas handling system is visible (white), cooling samples to temperatures below 20 mT.

When implanted into matter, the positive muon acts as a microscopic local probe, which sensitively interacts with the characteristic electronic and magnetic environment. This instrument is uniquely suitable for the study of unconventional and high-T_c superconductivity. An example is shown in Figure 3. It can map internal magnetic fields induced by super-currents, detect magnetic, superconducting and non-magnetic phases simultaneously, and follow their relevant dynamical fluctuations. For the high-temperature superconducting materials, this suitability is even more accentuated because the underlying mechanisms responsible for superconductivity are themselves believed to be magnetic. Also magnetic order (e.g. spin density waves) eventually present in modulated superconducting phases with mixed order and coupling to it may be studied. The world's sole necessary combination of very low temperatures (20 mK) and high magnetic field is particularly important for studies of quantum phase transitions (QPT) and of critical points in magnetic and superconducting materials. Systems of topical interest are those that have frustrated interactions or strong quantum fluctuations such as quantum magnets and that exhibit "spin liquid" or other complex ground states. A prominent QPT such as the one associated with Bose-Einstein Condensation should be detectable by a spin probe. Finally, the very good time resolution will allow extending the range of observable spin relaxation rates for instance in molecular magnets, where spectral information and tunneling rates between discrete levels can be extracted by measurements in high magnetic fields.

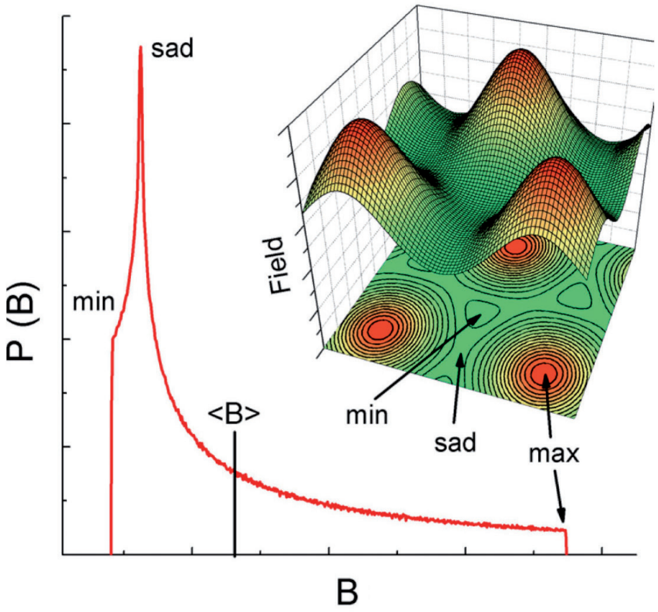

Figure 3: Field map of a hexagonal vortex lattice in an ideal extreme type-II superconductor. A high-field µSR experiment measures the magnetic field distribution; fundamental length scales characterizing the superconductor are then extracted.

Resonant inelastic-X-ray scattering RIXS

Resonant inelastic X-ray scattering (RIXS) is a powerful spectroscopy technique, which is atomic site selective. This is due to the involvement of a core-hole in the intermediate state created by resonant excitation to the conduction band. With RIXS the electronic structure of condensed matter is probed by determining the energy and symmetry of charge neutral electronic excitations (e.g. crystal field, charge transfer, phonon or spin excitations) in strongly correlated materials such as transition metal oxides and rare earth systems. Variation of the scattering angle can be used in RIXS to study the dispersion of these excitations as a function of momentum transfer, thereby enabling characterization of their localized vs. delocalized character.

The SAXES instrument at the ADRESS beamline of the SLS (see Figure 4) is a high-resolution X-ray emission spectrometer following an optical scheme based on two variable line spacing (VLS) spherical gratings with radius of 58.55 m and 60 m for an average groove density of 3200 lines/mm and 1500 lines/mm, respectively. With an input arm around 1 m and exit arm of up to 4 m, SAXES utilizes a total length of 5 m. The detector used is a liquid nitrogen cooled two-dimensional CCD camera with a pixel size of 13.5 µm and effective spatial resolution of about 24 µm. The long dispersion arm and advanced detector enable extremely high resolving power of better than 10'000 below 1100 eV, in particular 17'000 at the Ti L_3 edge (470 eV) and 12'000 at the Cu L_3 edge (930 eV). The total operative energy range of SAXES is 400 – 1600 eV with a typical spot size on the sample below 5 µm.

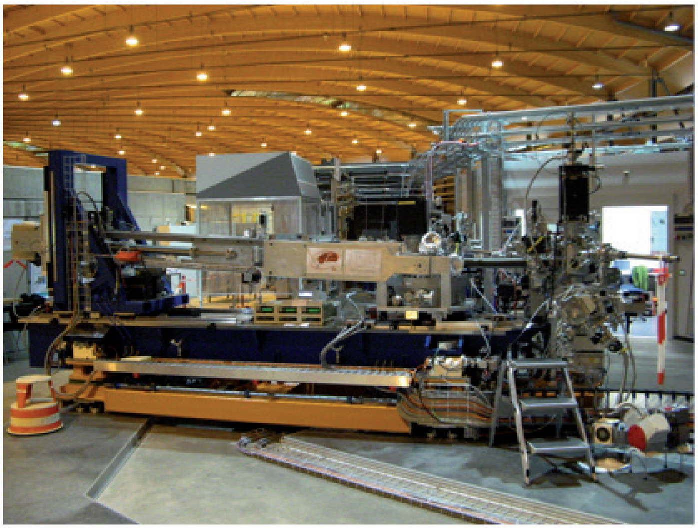

Figure 4: RIXS endstation at the ADRESS beamline of the SLS consisting of the analysis chamber (on the right), SAXES spectrometer and rotatable girder platform.

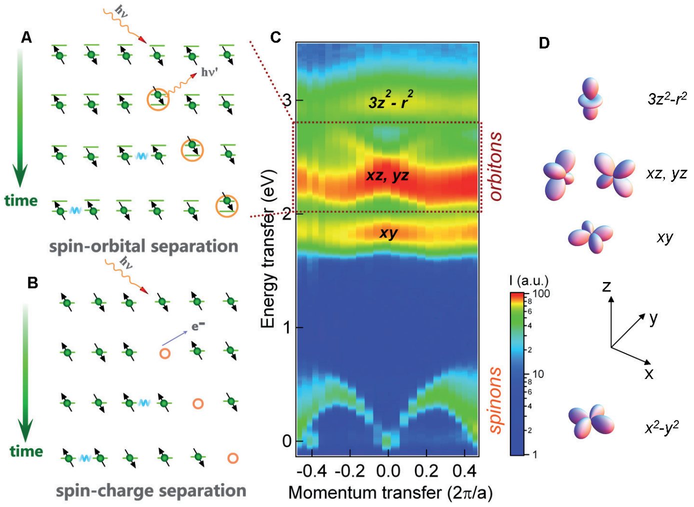

*Figure 5: Schematic illustration of (**A**) spin-orbital and (**B**) spin-charge separation processes. (**C**) RIXS intensity map of the dispersing spinon and orbiton excitations vs. photon momentum transfer along the chains and photon energy transfer. (**D**) Symmetry of the involved Cu 3d orbitals [Reproduced from J. Schlappa et al., Nature **485**, 82 (2012)].*

In a recent highlight the SAXES instrument was used to map momentum dispersive spectra from collective spin- as well as orbital- excitations in the spin-chain compound Sr_2CuO_3 with Cu L_3 edge RIXS (see Figure 5). From this study it was shown for the first time that in a one-dimensional spin system the electronic degree of freedom can split up in its spin and orbital parts. Another powerful demonstration of the high sensitivity and capabilities of the RIXS set-up at SLS is the detection of spin waves in isolated single atomic La_2CuO_4 layers.

Future experiments at the SwissFEL

The PSI has proposed the construction of a hard x-ray free-electron laser facility, the SwissFEL, which will produce intense, ultra-short pulses of coherent radiation in the energy range from 2 keV to 13 keV, with a future extension to the soft x-ray range (see Figure 6). The science case for the SwissFEL project emphasizes the dynamics of condensed matter systems, chemical systems and the damage-free imaging of nanostructures. The Swiss parliament has approved the proposal by the end of 2012. The preparation of the site has already started. The goal is to produce the first beam by fall 2016. Pilot experiments should start in spring 2017. The specifications of the experimental stations have been discussed with user groups in a series of workshops held at PSI. A Conceptual Design Report will be soon available (more information: http://www.psi.ch/swissfel/).

In order to prepare for SwissFEL and to start work in the exciting area of ultra-fast spectroscopy several teams from Swiss universities and PSI are using some of the few existing x-ray free electron lasers worldwide to do first experiments. Here, the ability to manipulate matter on ultra-short time scales offers potential breakthroughs in future device technologies as well as a better understanding of fundamental material properties. For this purpose, it is of immense importance to be able to selectively drive excitations of interest. A team of researchers from PSI, the ETH, Stanford (SLAC) and Berkley has recently demonstrated this with an experiment performed in July 2012 at the x-ray free electron laser LCLS (Linac Coherent Light Source at Stanford, US). They showed that a short THz-frequency pulse may be used to excite a coherent electromagnon in multiferroic $TbMnO_3$. Electromagnons are hybrid excitations of a magnon and a phonon that couple polarization with magnetism in multiferroic materials. The material response was studied by time dependent resonant magnetic soft x-ray diffraction. This experiment demonstrates that newly available methods of creating short, intense THz-frequency pulses make it possible to coherently control magnetic moments. X-ray free electron lasers allow us to study these excitations in real time. With the upcoming SwissFEL, studies of the detailed mechanism of switching electric polarization through selective excitations will become possible.

Conclusions

The complexity of materials requires a combination of probes and studies over a variety of length, energy, and time scales to be able to investigate their structural, electronic and magnetic properties. While these studies are of fundamental interest and challenge our understanding of the complex correlations and interactions that are at work in such systems, materials are essential for technological applications ranging from IT devices to energy harvesting and storage. With the investment in new neutron, µSR, and resonant x-ray spectrometers and the construction of the x-ray free electron laser SwissFEL the PSI enables new experimental approaches, which are available to the wide and diverse Swiss physics community.

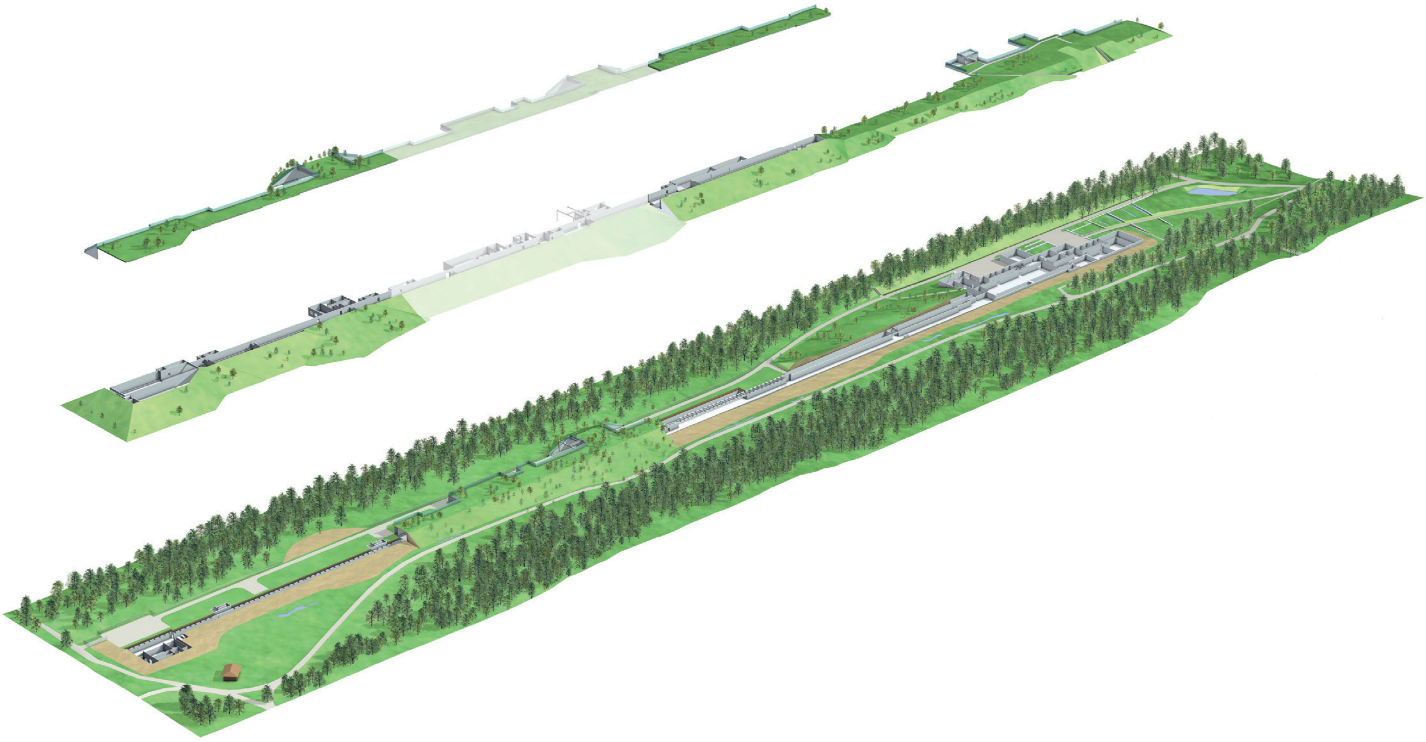

Figure 6: Layout of the future SwissFEL at PSI. Its total length is 700 m.