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New enhanced tool for neutron spin echo spectroscopy

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A restriction for any wave interference based scattering technique (*e.g.* neutron or X-ray diffraction, scattering or spectroscopy) is that such methods probe the Fourier transform of the space–time correlations. Such information, on its own, cannot be directly converted into an atom-by-atom movie of the structure and motion. Nevertheless, in the spirit of crystallography from its beginnings, scattering patterns can ultimately be 'solved' and translated into atom-by-atom structures in real space. With the support of model calculations, the inelastic neutron scattering spectra of the dispersion patterns of wave-like elementary excitations in crystalline matter can also be computationally transformed into atom-by-atom movies. For noncrystalline matter and disordered phenomena, the combination of neutron spectroscopy and molecular dynamics calculations offers increasingly powerful opportunities for translating diffuse scattering patterns into atomic level space–time domain information.

Neutron spin echo (NSE) spectroscopy offers unique capabilities within the spectrum of neutron scattering techniques by extending the range of the time domain accessible to spectroscopic exploration by one–three orders of magnitude compared to other neutron techniques. This is achieved by labeling the neutrons with their spin orientation, and then comparing the initial and final neutron velocities in a scattering experiment individually for each labeled neutron. Thus, changes in neutron velocity in the scattering event down to one part per million (p.p.m.) (directly reflecting the frequency spectrum of the atomic motion probed) can be observed for all neutrons in the beam, even though the initial neutron velocity is only defined to a precision of a few percent. Selecting the initial neutron velocity with 1 p.p.m. accuracy in a conventional spectroscopy experiment would prohibitively reduce the beam intensity (and there is no established way to do this anyway). Furthermore, the NSE velocity labeling method – based on neutron spin Larmor precession on passing through well defined magnetic fields – implies an automatic Fourier transformation of the velocity spectra. Thus, the physical time variable in NSE experiments is the one directly observed. For example, if we study the lifetime of an elementary excitation in condensed matter (phonon, magnon *etc.*), the directly observed data will show the decay of the excitation wave in real time. For more details see Mezei (1983), and references therein. The example in Fig. 1 (Keller *et al.*, 2006) shows the decay in time of high-frequency transverse sound wave phonon excitations in lead, directly revealing for the first time the impact of superconductivity on phonon lifetime.

The recent paper of Li & Pynn (2014) provides an impressive new tool for using NSE spectroscopy for the observation of the lifetime or frequency shift associated with elementary excitations in crystalline matter. The specific challenge in such work is that the frequency of an elementary excitation varies with its wavelength, so if the wavelength is not infinitely well defined, the variation of frequency over the selected, necessarily finite, wavelength range will mask the broadening of the frequency spectrum due to the finite lifetime. The NSE spin labeling technique can be extended so that it is also sensitive to the direction of neutron velocity, and it has been shown that this provides a handle to filter out (to first order) from the NSE response the effects related to the dispersion of the elementary excitation wave, *i.e.* the wavelength dependence of the frequency. Originally, this method was successfully demonstrated by using direct currents in magnetic field coils for fine-tuning the shape of the Larmor precession fields (see Mezei, 1983). But successful use of the NSE method for the study of the lifetime of elementary excitations in hard matter (see Mesot, 2006, and references therein) required a more powerful radio-

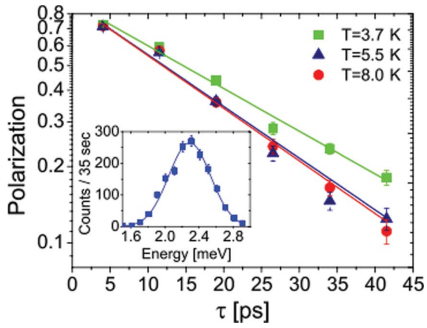

Figure 1
The NSE polarization signal of T1 transverse phonons in lead below and above its superconducting transition temperature of 7.2 K (Keller *et al.*, 2006). The inset shows the signal of the same phonons observed by conventional neutron triple-axis spectroscopy. The 25–30 ps decay lifetime shown by the NSE signal corresponds to about 0.04 meV linewidth in the energy spectrum in the inset. This small width is fully masked by the 0.6 meV intrinsic width of the triple-axis spectrometer resolution function, which dominates the signal in the inset. Courtesy of T. Keller *et al.* Copyright (2006) by The American Physical Society.

frequency (RF) spin modulation tool and a change in the configuration and orientation of the RF coils to accomplish the tuning of the effective field shape. Before this advance improved the resolution by about an order of magnitude, such fine information on elementary excitations was only obtainable in a few special cases.

The capability of using neutron spin labeling to encode information on the direction of the neutron velocity has also been developed into a standalone variant of the NSE method for very small angle neutron scattering studies of the nano- and mesoscale structure in materials up to the micrometre length scale (Pynn *et al.*, 2005, and references therein). Li and Pynn have now developed the use of superconducting direct current coils, acting as magnetic Wollaston prisms, to allow essentially unlimited tuning of NSE Larmor precession field shapes for fine-resolution studies of practically all elementary excitations accessible to neutron scattering observation.

The construction of the next-generation European Spallation Source (ESS) in Lund, Sweden, will open up new opportunities for neutron scattering by delivering up to two orders of magnitude brighter neutron beams to the sample. This will allow the observation of small signals from small samples that are not available in cubic centimetre quantities, the typical sample volume for current neutron scattering work. This gain will greatly benefit the use of NSE spectroscopy in the picosecond to microsecond time domain for the study of structural and magnetic relaxation phenomena. Here NSE only needs encoding of the absolute value of the neutron velocity (*i.e.* no encoding of neutron flight direction). The high neutron intensity capability of ESS is intrinsically based on its generation of pulsed neutron beams. Ironically for this comment, the fine-resolution study of elementary excitations by the NSE method is the only experimental technique in neutron scattering for which no way is yet known for efficiently taking advantage of pulsed neutron sources. This remains a next challenge for experimental ingenuity, of which the development of magnetic Wollaston prisms for NSE spectroscopy is a prime example.

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