LAGRANGE - the new neutron vibrational spectromter at the ILL

Mónica Jiménez-Ruiz, Alexander Ivanov and Stephane Fuard
Institut Laue-Langevin, 6 rue Jules-Horowitz, BP 156, F-38042, Grenoble Cedex
E-mail: jimenez@ill.fr

Abstract. The new neutron vibrational spectrometer LAGRANGE replaces the Be filter spectrometer placed on the hot source at the Institut Laue-Langevin. Lagrange provides much higher sensitivity to extremely small or low scattering samples through substantially increased detector count rate combined with considerably improved energy resolution. It opens new possibilities for the spectroscopic studies of complex materials. The new instrument permits investigation of much smaller samples, inherent in novel materials studies, recording of well-resolved vibration spectra. Lagrange is fully operational for users since 2012.

1. Introduction
Vibrational spectroscopic techniques provides unique information about the high-energy atomic and molecular vibrations in condensed matter systems. The neutron scattering spectroscopy has proven to be a powerful and unique tool for studies of dynamical properties of solids and associated physical and chemical phenomena. Neutrons probe the bulk properties of samples and can easily penetrate through container walls making possible studies of unstable or conditioned materials. The information obtained by Inelastic Neutron Scattering (INS) experiments is complementary to the one obtained by means of infrared and Raman spectroscopy that yield information about lattice vibrations, which produce a variation of the dipole moment or the polarizability of the system respectively. The INS method directly follows the motions of nuclei and, since there are not specific selection rules, the INS spectra provide information about all vibrations of the system. For this reason INS technique is used as an important source of supplementary data for computational techniques dealing with the dynamics of complex systems.

The Lagrange project was launched with the goal to create a new spectrometer for studies of lattice and molecular excitations in the extended energy range up to several-hundred-meV typical for materials containing light chemical elements, in particular hydrogen. Lagrange replaces the Be filter spectrometer placed on the hot neutron spectrometer IN1 at the Institute Laue-Langevin [1] and dedicated to the studies of molecular vibrations in complex systems, such carbon nanotubes [2], zeolites [3] and hydrides [4]. Another instrument dedicated to the molecular vibrational spectroscopy is the high-resolution crystal analyser spectrometer, TOSCA, at ISIS pulsed neutron source [5].

2. IN1-Lagrange design
Lagrange is placed on the hot neutron beamtube H8 at the Institut Laue-Langevin (ILL) and shares this position with the diffractometer for liquids and amorphous systems D4 and with...
the hot neutron Three-Axis spectrometer IN1. Since Lagrange shares the primary spectrometer with these instruments, it benefits from the recently upgraded double-focussing monochromator with a set of different reflecting planes:

- Cu(220) reflection, with an starting incident energy of $\sim 27$ meV up to $\sim 500$ meV, used for the high flux configuration,
- Cu(331) reflection, with an starting incident energy of $\sim 65$ meV up to $\sim 1$ V, used for the improved energy resolutions configuration at high energies, and
- Si(311) reflection, with an starting incident energy of $\sim 16.5$ meV up to $\sim 200$ meV; and the Si(111) reflection, with an starting incident energy of $\sim 5.5$ meV up to $\sim 20$ meV used for the low-energy part of the spectra and that present the advantage of suppressed second order contamination in the monochromatic beam.

With the present reflections, this monochromator supplies at present more than twice increased monochromatic flux in the high neutron energy range available from the hot neutron source of ILL reactor.

Figure 1. Lagrange design.

The LAGRANGE (LArge GRaphite ANalyser for Genuine Excitations) secondary spectrometer setup (Fig. 1) is based on the space focussing of neutrons scattered by the sample in a very large solid angle, all of them registered with a relatively small single counter (a He3 gas detector). The focussing reflecting surface of $1 \text{ m}^2$ is built around the vertical sample-detector axis from pyrolytic graphite crystals set to reflect neutrons with the fixed average energy of 4.5 meV. The appropriately shaped cooled Beryllium filter is installed right after the sample in order to remove higher-order harmonics in the analyser reflections. It is important that the absolute value of the accepted solid angle ($\sim 2.5$ steradian) stays among the highest of the most ambitious instrument projects at present. At the same time it is realised in the relatively small characteristic spectrometer volume. The high-energy resolution of the new instrument is attained with a relatively low grade of the pyrolytic graphite. The carefully designed screen of boron-containing absorber is installed on the sample-detector axis in order
to suppress the intense elastic scattering from the sample. Further reduction of the instrument background, contaminated with the high neutron energy components, is achieved through a massive polyethylene shielding built around the whole analyser. Both the sample cryostat and the filter cooling circuits are equipped with powerful closed-cycle helium refrigerators. Note that there are no mutually moving parts within the secondary spectrometer which is positioned as a whole around the monochromator in order to record the inelastic scattering spectra changing step-by-step the incident energy similarly to a typical experiment on a three-axis spectrometer.

3. Results
Lagrange has been firstly commissioned during the 2nd reactor cycle of the year 2011. The first inelastic scattering spectrum measured with Lagrange is presented on the Fig.2. We used a sample of 2.5-diiodothiophene, \((\text{CH})_2(\text{Cl})_2\text{S}\), for characterisation of the instrument performance. This material is known to exhibit very sharp vibration lines at low temperatures so that it can be used as a standard for measurements of the spectrometer resolution. The visible sharp peaks in the energy spectrum on Fig.2 are due to vibrations involving hydrogen atoms, present in the sample in a relatively small quantity: 6 milligram of the partial hydrogen weight. It should be noticed that the collected statistics in the energy interval below 200 meV correspond to some 12-30 seconds of the measuring time per point (the difference is due to energy variation of the incident monochromatic neutron flux) so that this informative energy interval could be scanned in less than 2 hours while the whole scan time for the shown spectrum, up to 600 meV, is about 4 hours.

![Figure 2. The inelastic scattering spectrum recorded with the Lagrange analyser and the 2D-focusing Cu220 monochromator. The sample of 2.5-Diiodotiophene is cooled down to the base cryostat temperature of 2.5 K. The intensity is as-measured, without any further treatment, no background (cryostat and sample holder) is subtracted.](image)

The energy resolution of Lagrange can be further improved in the broad energy range using of the available Cu331 reflecting plane of the IN1 monochromator, as it is seen from the spectra presented on the Fig.3. Many more fine details of the vibration density of states are revealed in the higher resolution run in the expense of solely \(\sim 2\)-fold decrease in the statistics at the same counting time.

The measured line-widths with the 2.5-diiodothiophene sample are presented as points in the Fig.4 which also shows calculated energy resolution curves with different analyser and monochromator settings. We note that similarly to the predictions of the intensity gain,
Figure 3. Inelastic scattering spectra (the same sample as on the Figs. 2) measured with different monochromator planes, Cu220 and Cu331. No background is subtracted. The counting time per point was about 20–30% higher in the case of Cu220 monochromator what corresponds to about a factor of 2 in the intensity decrease at the same counting time for the high-resolution setup.

Figure 4. Calculated (lines) and measured (points) relative energy resolution of Lagrange with different primary spectrometer settings. The black line corresponds to the Be-Filter analyser resolution with the Cu220 monochromator, blue line the same in the case of Lagrange, red line Lagrange with the Cu331 monochromator. These calculations are made for the natural collimation of the in-pile beam tube of IN1 and 20’ Soller collimator in the monochromatic beam. Points are the peak widths divided by the peak energies as measured in the corresponding conditions with the 2.5-diiodothiophene sample. The lowest broken line represents the best-expected future energy resolution of Lagrange.

The resolution performance is in full accord with what was expected from the pre-project estimations. The best expected energy resolution, of the order of 1% in the most informative energy range 100-200meV, should be achieved with already manufactured 12 and 10 Soller collimators installed before and after monochromator. This ultimately high energy resolution, will bring Lagrange to the top of the high-resolution neutron spectrometers elsewhere and will be available with the future collimator changer installed before the monochromator. The changing over different resolution settings can be very easily done leaving users the flexibility to optimise their measurements with respect to resolution vs intensity choice.
4. Conclusions
The new neutron vibrational spectrometer LAGRANGE has proved as a highly sensitive and a high-resolution instrument. It opens new possibilities for the spectroscopic studies of complex materials. The new instrument permits investigation of much smaller samples, inherent in novel materials studies, recording of well-resolved vibration spectra, a more detailed probing of external parameters like temperature, pressure, chemical composition, magnetic field, time-dependent processes etc. It makes Lagrage highly competitive with the instruments for high-energy neutron spectroscopy elsewhere.

The new instrument provide much higher sensitivity to extremely small or low scattering samples through substantially increased detector count rate combined with considerably improved energy resolution in comparison to the beryllium filter-analyser used in the past on IN1. Lagrange is fully operational for users since 2012 and several improvements are in progress on sample environment (gas injection, heating insert and pressure cells).

References
[1] http://www.ill.eu/instruments-support/instruments-groups/instruments/in1-taslagrange/characteristics/
[2] Rols S, Benes Z, Angleret E, Sauvajol JL, Papanek P, Fischer JE, Coddens G, Schober H and Dianous AJ 2000 Phys. Rev. Lett. 85 5222.
[3] Jobic H 2002 Current Opinion Sol. State and Mat. Science 6 415.
[4] Kolesnikov A et al 2002 Physica B 316-317 158-161.
[5] Parker S 2002 J. Neutron Res. 10 173.