JANUS – INS Instrument for Catalysis

JANUS is a medium-high resolution, broadband, indirect geometry spectrometer coupled together with a medium resolution direct geometry spectrometer. JANUS is optimized for materials chemistry and catalytic studies involving hydrogenous materials.

Science Drivers

Inelastic neutron scattering (INS) provides insight into the microscopic dynamics of molecular species. Neutrons are uniquely sensitive to hydrogen and hydrogenous species due to the large incoherent cross section of hydrogen. Both direct and indirect geometry spectrometers have a role in understanding the dynamics of hydrogen in catalytic reactions. Direct geometry instruments like ARCS and SEQUOIA typically provide good Q-resolution while indirect geometry instruments like VISION often integrate over Q in order to get high sensitivity and count rates. Particularly in the case of hydrogen, maintaining good Q-resolution at low scattering angles can provide more detailed information on dynamics that is not degraded by the presence of vibrational overtones and combinations. JANUS combines both approaches into a single instrument, alternately providing good Q-resolution at low scattering angles as a direct geometry spectrometer or maintaining the high sensitivity and count rate at higher scattering angles by integrating over Q like VISION does. An instrument that can do each of these functions greatly facilitates the study of irreversible phenomena and in situ manipulation of samples. JANUS will address topics where the dynamics of hydrogen is of particular relevance including:

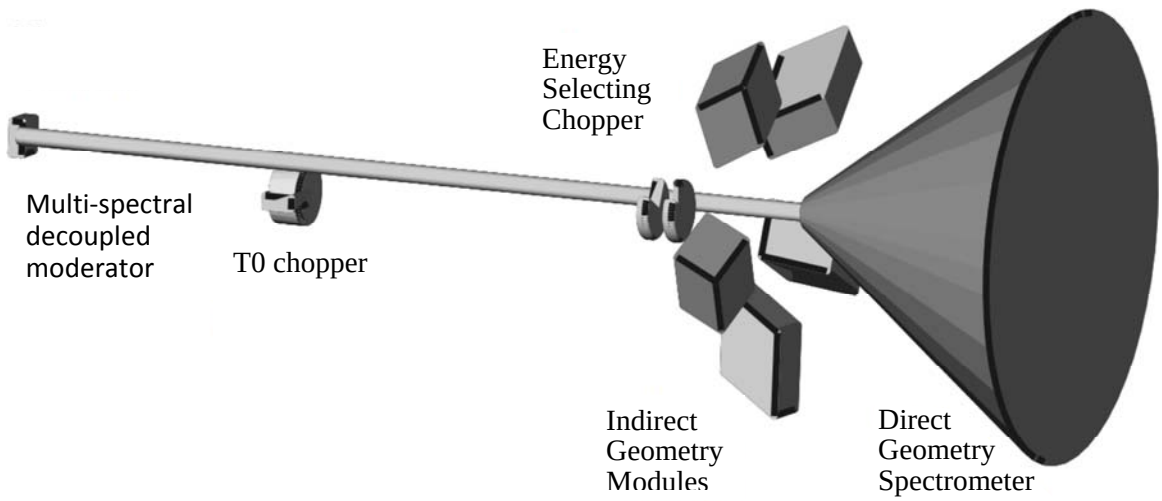
- Catalysis: Much of catalysis is concerned with the transfer of hydrogen between reagents and products. Access to the C–H/N–H/O–H stretch region provides a very simple means of identification and is ideally suited to quantification of the species present. The 0–2000 cm^{-1} region is where the details of the catalyst–adsorbate interaction can be discerned and the indirect backscattering detectors provide the good energy resolution needed for this. However, the surface species of interest are often present in low concentration, and good resolution is of no use if there is insufficient sensitivity. This problem is addressed by the direct forward scattering which will provide at least a tenfold increase in sensitivity and possibly as much as a hundredfold increase over the backscattering detectors. The improved sensitivity will also allow industrially important non-hydrogenous adsorbates, such as CO_x, NO_x and SO_x, to be studied for the first time.
- Proton conductors are heavy metal oxides that are hydrated to generate mobile hydroxyls and have considerable potential in intermediate temperature fuel cells. These materials exhibit multiple sites for incorporation of hydroxyls; thus, diffraction studies are only able to provide an average picture of the material, and the crucially important local structure around the hydroxyls is lost. Spectroscopy provides this missing local picture, and INS (in conjunction with first principles modeling) is particularly matched to these needs as the low energy modes enable discrimination between the various possible sites. These modes are often weak or invisible to optical probes. In addition, access to the O–H stretch region is needed to quantify the species present and to discriminate between hydroxyls and water.

Science Requirements

1. An inelastic spectrometer that can provide good Q resolution at low scattering angles ≤ ±30 deg and the ability to provide high count rates and sensitivity by integrating over Q at higher scattering angles.
2. Large dynamic range of energy transfer from -3.0 meV to 500 meV in indirect geometry mode; incident energies up to 300 meV in direct geometry mode.
3. Energy resolution: 1% $\Delta\omega/\omega$; ~150 μeV (FWHM) at elastic line in indirect geometry mode
4. Integrated suite of sample environment equipment that provides extreme sample conditions in high pressure, low temperature, and high magnetic field).

Technical Description

This instrument will nominally view 7 x 7 cm^2 water face of the multispectral decoupled moderator. At 20 m from the moderator, JANUS can use neutron wavelengths as long as 19.8 Å in the first frame, covering a wide range of energy transfers simultaneously in indirect geometry operation. JANUS cannot run in both direct and indirect geometry modes of operation simultaneously so the energy selecting choppers directly in front of the sample will need to be parked open when in indirect geometry operation. JANUS will have at least one T0 chopper to eliminate the prompt pulse and bandwidth choppers to provide sharp definition of the wavelength band incident on the sample when in indirect geometry mode. The figure below gives a schematic view of JANUS.



Key parameters of JANUS

Parameter	Description
Moderator	7 x 7 cm^2 water face of the multi-spectral decoupled moderator
Beam size	1×1 cm^2
Moderator–sample distance	20 m
Wavelength range	0.4 Å ≤ λ ≤ 19.8 Å
Resolution	indirect geometry: 150 μeV (FWHM) at the elastic peak; $\Delta\omega/\omega$ ~1% direct geometry: $\Delta E_{incident}/E_{incident}$ ~1-2%
Detector	linear position-sensitive ^3He