Applications

- To characterise interaction energies associated with bonding of atoms in solids and liquids
- To measure the energy level spacing in magnetic ions and of the interactions between them
- To observe the interaction of magnetic moments with their surrounding ions (crystal fields) or with conduction electrons (fluctuating valence, heavy fermions)
- To determine vibrational states in amorphous solids and polycrystals
- To examine molecular excitations in various materials, also those of technological interest (zeolites) and especially in diluted systems (matrix isolation)

Selected examples

Characterisation of the ligand-field splitting of Fe(II) for a biologically relevant ligand.


The imidazole ring, as a histidine moiety, functions as a ligand towards transition metal ions in a number of biologically important molecules. In particular, it is found coordinated to iron in haemoglobin, myoglobin, and cytochrome c. The majority of previous studies on iron(II)-imidazole complexes have focused on model systems, resembling the naturally occurring low-symmetry environments, which are necessarily so complicated that the role of the individual ligands is difficult to determine. The approach of this work is to study high-symmetry homoleptic complexes, in order to obtain an understanding of the iron-imidazole interaction by the determination of the ground-state electronic structure. A direct spectroscopic determination of the electronic structure can be obtained using inelastic neutron scattering (INS), and IN4 is ideally suited.

The ligand-field in this case has both a spin-orbit coupling and a symmetry-imposed trigonal field contribution; by directly probing the electronic states using INS one can characterise the effect of imidazole ligation to the metal. The ground-state to first excited-state transition at ca. 2 meV \( \left( \text{I} \right) \) had been previously observed on FOCUS at PSI, alluding to the presence of a hot-band at ca. 9 meV \( \left( \text{II} \right) \) on the basis of existing models of ligand-field splitting of the \( ^{5}T_{2g} \ (O_{h}) \) ground term.

IN4 data collected on a 5 g sample of the fully-deuterated nitrate salt clearly shows the increase in intensity of both \( \text{I}' \) (the induced depopulation of the excited state) and \( \text{II} \) as a function of temperature, with a concurrent decrease in \( \text{I} \) due to thermal depopulation of the ground-state. The \( Q \)-dependence of these features is consistent with the free-ion form factor of Fe(II) and is easily discriminated from the low-lying phonons growing up around 5 meV.

This experimental result provides a complete characterisation of the ground-term splitting of imidazole ligation to Fe(II) for the first time. In addition, by using atomic overlap model bonding parameters, it will be applicable to lower-symmetry transition metal sites in biologically active systems.
IN 4 is a time-of-flight spectrometer used for the study of excitations in condensed matter. It works in the thermal neutron energy range (10 ... 100 meV).

Instrument description

Primary spectrometer

The main components of the incident beam part of the instrument are the two background choppers, the double curvature monochromator with four faces and the Fermi chopper.

The background choppers are rapidly pulsating beam shutters which act as a low bandpass filter. Thus they eliminate from the beam most of the fast neutrons and gamma rays that would give background noise in the spectra. The modular shielding encloses the background choppers in separate compartments in order to cut off these undesired neutrons as early as possible.

A suitable energy is selected from the thermal neutron spectrum with the crystal monochromator. The monochromator, an assembly of 55 crystal pieces, concentrates the divergent incident beam onto a small area at the sample position. The full use of the available solid angle gives a high incident flux. The variable curvature of the monochromator is essential in controlling the time and space focusing conditions for optimal performance (see H. Mutka, Nucl. Instr. and Meth. A 338 (1994) 144).

The Fermi chopper rotates at speeds of up to 32000 rpm. It transmits short neutron pulses (10 ... 50 µs) to the sample. The time-of-flight of neutrons between the chopper and the sample (1 ... 5 ms) can be measured by using precise electronic circuitry.

Secondary spectrometer

The sample environment is designed to accommodate standard cryostats and furnaces. A radial collimator around the sample position is used to reduce the scattering from the sample environment.

The secondary flight-path is in vacuum to avoid parasitic scattering of the transmitted neutrons.

The detector bank covers scattering angles of up to 120°. In addition to the 3He detector tubes (length 300 mm, width 30 mm, elliptical section, pressure 6 bar) a 3He filled multidetector (eight sectors with 12 radial cells each; outer diameter 60 cm) will allow us to observe forward scattering.

The time-of-flight spectra measured at various angles are further treated in order to obtain the scattering function $S(Q,w)$ that is characteristic of the properties of the sample. One can extract from it the complete information on the energy and length scale of the dynamical phenomena of isotropic systems such as liquids, disordered materials as well as of non-dispersive (i.e. localised) excitations in molecules and magnetic ions. Otherwise orientationally averaged quantities are obtained, for example the density of vibrational states in polycrystals.

Instrument layout

Instrument Data

Reactor hall, thermal beam H12

<table>
<thead>
<tr>
<th>monochromator</th>
<th>take-off angle $2\theta_t$</th>
<th>resolution $\Delta E/E_i$</th>
<th>flux on sample</th>
<th>background choppers $\nu$ max.</th>
<th>Fermi chopper $\nu$ max.</th>
<th>duty cycle</th>
<th>beam size</th>
<th>primary collimation $\Delta 0$</th>
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</thead>
<tbody>
<tr>
<td>PG 002: 2.00 ... 3.8 Å</td>
<td>35 ° ... 70°</td>
<td>2 ... 5 %</td>
<td>$5 \times 10^4$ n cm$^{-2}$ s$^{-1}$</td>
<td>5000 rpm</td>
<td>40000 rpm</td>
<td>3 x 100</td>
<td>3 x 8 cm$^{-2}$</td>
<td>1°</td>
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<tr>
<td>PG 004: 1.00 ... 1.9 Å</td>
<td>Cu 111: 1.25 ... 2.4 Å</td>
<td>Cu 220: 0.80 ... 1.5 Å</td>
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