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The very low angle detector for high-energy inelastic neutron scattering on the VESUVIO spectrometer

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Abstract

The Very Low Angle Detector (VLAD) bank has been installed on the VESUVIO spectrometer at the ISIS spallation neutron source. The new device allows for high-energy inelastic neutron scattering measurements, at energies above 1 eV, maintaining the wave vector transfer lower than $10\,\text{Å}^{-1}$. This opens a still unexplored region of the kinematical (q,ω) space, enabling new and challenging experimental investigations in condensed matter. This paper describes the main instrumental features of the VLAD device, including instrument design, detector response, and calibration procedure.

Keywords: Neutron scattering; Neutron instrumentation; HINS

1. Introduction

The advent of the pulsed neutron sources has opened up new fields of investigation in condensed matter, as the intense fluxes of epithermal neutrons (above 500 meV) allow to achieve energy, ω, and wave vector, q, transfers unaccessible at steady state reactors. The investigation of condensed matter at small scales of time ($\leqslant 10^{-15}$ s) and distances ($\leqslant 1\,\text{Å}^{-1}$) is very attractive, as information on the single particle dynamics, namely atomic momentum distribution $n(p)$ and mean kinetic energy $\langle E_K \rangle$, can directly be accessed through the deep inelastic neutron scattering (DINS) technique [1–3]. On the other hand, epithermal neutrons allow to access the kinematical region of the (q, ω) space characterized by $\omega \geqslant 1$ eV and $q \leqslant 10\,\text{Å}^{-1}$, i.e. the high energy inelastic neutron scattering (HINS) regime [4,5]. HINS enables experimental studies on the dispersion relations of high energy excitations in metals, semiconductors and insulators, high lying molecular rotational–vibrational states, molecular electronic excitations, and the electronic level in solids [6–9].

The most effective way to fulfill the HINS kinematical conditions is to use the inverse geometry configuration, detecting scattered neutrons well above 1 eV at small angles [10–12]. To this aim, the experimental concept of the Resonance Detector (RD), proposed in the late 70s [13,14], has been revisited and optimized for the VESUVIO beam line [15,16] at the ISIS spallation neutron source [17]. Different types of photon detectors have been successfully tested on VESUVIO in the RD configuration for DINS measurements, at scattering angles above 20° [18–20]. Their use at the very small scattering angles ($\leqslant 5°$) required for high ω and low q was a more recent development

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and required structural modifications of the original spectrometer layout. The beam line of the instrument was modified by tightening the incident neutron collimation, and by fitting a new vacuum tank which provides minimum attenuation of the neutrons scattered at low angles. A prototype of the VLAD bank was previously constructed and successfully tested with benchmark HINS measurements [4].

This paper concerns the main features of the final VLAD device, including instrument design, detector response, and calibration procedure. Section 2 provides a brief overview of the new scientific opportunities opened by this instrument development, while Section 3 discusses the kinematical requirements of HINS on an inverse geometry time of flight spectrometer. Sections 4 and 5 provide a description of the system and its calibration. Some concluding remarks are made in Section 6.

2. New science requirements

Accessing the HINS regime will allow for new experimental studies in condensed matter. HINS provides information on the dynamical structure factor $S(\vec{q},\omega)$ on the nuclear interaction of neutrons with atomic nuclei. $S(\vec{q},\omega)$ depends on the density of states $g(\omega)$ and on the vibration amplitude $\langle u^2\rangle$ of each atom in the molecule, weighted by its neutron scattering cross-section. In a hydrogen containing sample, the large incoherent cross-sections and the low mass of the proton ensure that the contribution to $S(\vec{q},\omega)$ coming from molecular vibrations involving hydrogen dominates the scattering intensity. In the kinematical region at low wave vector and high energy transfers (HETs), HINS allows access to the one-phonon component, $S_{m,\pm1}(\vec{q},\omega)$, of hydrogen-projected scattering function, $S_m(\vec{q},\omega)$. The expression of the dynamical structure factor in this case is given, in the isotropic approximation, by [21]

$$S_{m,\pm1}(\vec{q},\omega) = \frac{\hbar q^2}{4m}\exp\left(-\frac{1}{3}q^2\langle u^2\rangle_m\right) \times \frac{g(\omega)}{\omega}\left[\coth\left(\frac{\omega}{2k_{\mathrm{B}}T}\right)+1\right] \tag{1}$$

where $g(\omega)$ is the density of states projected on the hydrogen atoms of the unit cell and $\langle u^2\rangle_m$ is the nuclear mean squared displacement averaged over the hydrogen atoms of the unit cell.

On the other hand, HINS can be induced by the neutron magnetic interaction (through its magnetic moment) with the electron spin of the unpaired electrons. In such cases the magnetic interaction between neutron and electrons is described by the Hamiltonian:

$$H_{\mathrm{int}} = -\vec{\mu}_{\mathrm{n}}\cdot\vec{B} \tag{2}$$

where $\vec{\mu}_{\mathrm{n}} = -\gamma\mu_{\mathrm{N}}\vec{\sigma}$ is the neutron magnetic moment ($\vec{\sigma}$ being the Pauli spin operator acting on the neutron wave function, μ_{N} the nuclear magneton and $\gamma = 1.913$) and $\vec{B}$ the magnetic field of all electrons [22–26]. Consider a scattering process in which the system undergoes a transition from an initial state at energy E_{i} to a final state of energy E_{f}, while the neutron is scattered from an initial state of wave vector $\vec{k}_0$ and energy E_0 to a final state of wave vector $\vec{k}_1$ and energy E_1. In the first Born approximation, the double differential scattering cross-section is given by [25]

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E_0} = \frac{k_1}{k_0}|f(\vec{q})|^2\delta(E_{\mathrm{i}}-E_{\mathrm{f}}-\omega) \tag{3}$$

where $\vec{q} = \vec{k}_0 - \vec{k}_1$, $\omega = E_0 - E_1$ and $f(\vec{q})$ is the magnetic form factor. The latter is proportional to the classical electron radius $r_0 = e/4\pi\varepsilon_0 m_e c^2$, so that considering Eq. (3) the double differential scattering cross-section becomes proportional to r_0^2. Thus, as compared to the nuclear scattering cross-sections typical of DINS processes (ranging from few barns as for helium isotopes, to several tens of barns for hydrogen), those for magnetic interactions are at least one order of magnitude smaller.

Extending the energy transfer scale above 800 meV–1 eV with an associated low value of q, would allow a significant advance in the study of crystal field levels in rare earth intermetallic compounds and magnetic materials. For experiments involving still larger excitation energies (2–6 eV), epithermal neutrons can be used as a direct probe of the electronic band structure of materials such as semiconductor or insulators. On the HET spectrometer at ISIS, for example, inter-multiplet transitions with energies up to about 800 meV have been observed in Praseodymium [27] employing monochromatic incident neutrons up to about 1.7 eV. In order to observe higher lying transitions (e.g. the $^3H_4 \rightarrow {}^1G_4$ expected at about 1.2 eV) higher incident and final neutron energies are required, as briefly clarified in the next section.

3. Kinematical considerations

From a kinematical point of view, HINS requires that high energy neutrons in the final state be detected at small scattering angles. This can be argued by Fig. 1, where a contour plot of iso-q loci as a function of final energy and energy transfer ω is shown for three different scattering angles. Indeed, HINS provides access to a region of the (q,ω) space which is complementary to that of direct geometry chopper spectrometers as well as to other experimental techniques using different probes (see Fig. 2). As a specific example, the HET direct geometry time of flight spectrometer is optimized for high energy magnetic excitations above 50 meV, but being a direct geometry instrument the maximum energy loss is limited to about 2 eV.

For an inverse geometry instrument, where the scattered neutron energy is selected, there is no kinematical limit to the energy loss [28] and high ω can be achieved at low q values.