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Wide-angle polarization analysis on the multi-axis crystal spectrometer for the study of collective and single particle dynamics of methanol at its prepeak

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ABSTRACT

Neutron polarization analysis capability with a large scattering angle and momentum transfer coverage has not been widely implemented for the separation of coherent and incoherent quasi-elastic neutron scattering (QENS) spectra. A feasibility test for soft condensed matter applications was performed on the Multi-Axis Crystal Spectrometer (MACS) at the NIST Center for Neutron Research (NCNR). This capability benefitted from the high flux wide angle polarization analysis capability recently developed on MACS for hard condensed matter investigations. The experimental procedure is thoroughly described on how to take data and perform polarization efficiency correction before further data analysis is done. This capability was employed to unambiguously separate the coherent and incoherent signal from the QENS spectra for the investigation of both the collective and the single particle dynamics in partially deuterated (CH₃OD, D 99.5% purity) methanol. By studying a Q range encompassing the prepeak position, it was possible to directly measure the dynamics of the h-bonding associates which originate the prepeak signal, without contamination from the incoherent scattering, in spite of the relative weakness of the coherent scattering.

1. Introduction

Quasi-elastic Neutron Scattering (QENS) [1] techniques determine the energy exchanged between the incoming neutrons and the sample during the scattering process yielding a broadening of the elastic line, thus providing information on the relaxational processes in the sample. In state of the art neutron scattering spectrometers, energy exchanges between the sample and the neutron as small as $\approx 1 \text{ neV}$ can be appreciated (keep in mind that the energy of the incoming neutrons is of the order of the meV). Therefore QENS provides information on the lengthscales and timescales of motions over a range from a fraction of 1 Å to tens of Å and from a fraction of 1 ps to hundreds of nanoseconds. Such molecular insight is uniquely provided by neutron scattering as no other scattering methods access the above mentioned range. The neutron scattering signal can be divided into a coherent contribution, which depends on the relative position and motions of the atoms in the sample, and an incoherent contribution which only depends on the single particle dynamics of the atoms having a non zero incoherent

scattering cross section. Incoherent scattering originates from a random variability of the scattering length of the nuclei of the same atom; such variance can be due to the presence of different isotopes (isotopic incoherence) or by the existence of a nuclear spin, whose relative orientation with respect to the neutron spin affects the scattering length (nuclear spin incoherence). For example hydrogen has a very high (nuclear spin) incoherent scattering cross section, thus, the single particle dynamics of the hydrogen atoms has been extensively studied in liquids where the incoherent contribution is dominant [2,3]. Also the collective dynamics of some perdeuterated liquids was studied at Q values (Q being the exchanged wavevector) where the coherent scattering signal is significantly larger than the incoherent one [4–7], most commonly at the first sharp diffraction peak (FSDP) [8], yielding information on the structural relaxation. However, as the interest turns to increasingly more complex systems, the use of partially deuterated samples and the need to investigate the relative motion of specific structures has grown. In these cases at the Q values of interest, the signal can turn out to be a mixture of coherent and incoherent neutron

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scattering contributions which cannot be separated through usual methods and hardly through modeling efforts. The use of isotope substitution techniques may sometimes offer a way around the problem, as in the case of m-toluidine [5], methanol [7], and glyceline [9], but the availability of suitable deuteration schemes limits this option. Moreover, in some cases, especially with partially deuterated samples, coherent scattering peaks can contaminate the incoherent signal and affect the investigation of the single particle dynamics [10,11].

Neutrons possess an intrinsic magnetic moment of 1/2 h, called spin; this is a quantum property of the neutron and only two possible states can be measured $+1/2 \hbar$ and $-1/2 \hbar$. The use of polarized neutron beams and neutron spin analysis devices allows the separation of the coherent and incoherent scattering contributions [12,13]. In fact, coherent (as well as isotope incoherent) scattering does not alter the neutron spin whereas nuclear spin incoherent scattering has a 2/3 probability of flipping the spin of the scattered neutrons [12]. Thus, performing two measurements with the incoming beam polarized in two opposite directions and using an analyzer which allows detection of only one spin state, the fraction of coherent and incoherent scattering can be determined. Polarized neutron techniques have been extensively employed for the study of magnetism (magnetic scattering is mainly induced by the neutron's interaction with the dipolar magnetic moment from unpaired atomic electrons in the sample, a process accompanied by specific changes of the neutron spin state) especially on triple axis spectrometers; however, the application of polarized neutron techniques for the separation of coherent and incoherent nuclear scattering is limited, and, with few notable exceptions [10,11], more so in the case of QENS. Certainly this is due to a lack of suitable instruments which allow polarization analysis over a large scattering angle sector, in combination with a relatively large Q acceptance. Noticeable exceptions are the polarized diffractometers D7 at ILL (Institut Laue-Langevin) and DNS at JCNS (Jülich Center for Neutron Science), which offer a time-of-flight option [14,15], and more recently for the spectrometer LET [16] at the ISIS neutron and muon source of the Science and Technology Facilities Council (STFC) Rutherford Appleton Laboratory (RAL) [17].

Methanol is one of the most extensively studied liquids. This is due to its importance in many applications and because of its relevance as a model system ought to its molecular structure. In fact, with a hydrophilic hydroxyl group and a hydrophobic methyl group, methanol can be considered the simplest surfactant. It has also been employed as a simplified model system to investigate water, where one hydroxyl group has been substituted with a methyl one. Moreover, methanol is the first of the monohydroxy alcohol family. In these liquids, hydrogen bonding plays a fundamental role in determining their properties. In fact, in many instances they form molecular associates, most likely in the form of strings, through hydrogen bonding of their hydroxyl groups. Besides many other evidences, these associates are revealed by a prepeak in the structure factor as measured by x-ray or neutron diffraction. A prepeak is a feature located at Q values smaller than the FSDP, which thus indicates structuring at length scales larger than the first neighbor shell. The existence of a prepeak in methanol was signaled by x-ray diffraction at Q $\approx 0.8 \text{ Å}^{-1}$ [18] and through Molecular Dynamic (MD) simulations at $Q \approx 1.1 \text{ Å}^{-1}$ [19]. In a recent publication one of the authors has been able, with his collaborators, to experimentally highlight the existence of a prepeak in methanol at $Q \approx 1.1 \text{ Å}^{-1}$, using polarized neutron diffraction and a partially deuterated sample of methanol, CH₃OD [7]. The use of a partially deuterated sample is necessary; because of the negative coherent scattering length of hydrogen, in CH₃OD all the partial structure factors associated with the prepeak contribute positively whereas in the perdeuterated sample some of the partial structure factors are negative so that the prepeak signal is canceled out. However, without polarization analysis the prepeak signal would still be very difficult to observe because the incoherent scattering from the hydrogen atoms is the dominant contribution to the scattering [7]. Therefore, the dynamics of the molecular associates was investigated combining the data collected on four different methanol samples with different isotopic labeling. This method allowed the determination of the partial dynamics structure factor corresponding to the correlations between the hydroxyl and methyl hydrogens. The results indicate that the Transient Chain Model (TCM) [20] can be applied to methanol dynamics. Moreover, it was shown that the associate dynamics contributed in determining the methanol macroscopic viscosity [21].

The present contribution reports the results of the investigation of the dynamics of methanol using the cold Multi-Axis Crystal Spectrometer (MACS) at the NIST Center for Neutron Resaerch [22], with ³He neutron spin filter based wide-angle polarization analysis capability. Measurements were carried out on CH_3OD in a Q range encompassing the prepeak. The use of such an instrument allowed the separation of the coherent and incoherent scattering contributions, therefore allowing to determine the single particle dynamics of the hydrogen atoms in the methyl groups and the collective dynamics of the hydrogen bonded molecular associates.

In the next section of this paper the experimental details will be reviewed with special emphasis on the polarization analysis capability of the neutron instrumental setup. Afterward the data analysis method will be reviewed. The obtained results will be reported in the following section. This paper will conclude discussing the potentiality of polarization analysis methods for the study of soft matter systems in general and the opportunity and limitations offered by the MACS spectrometer.

2. Experimental setup

2.1. Sample

Partially deuterated (CH₃OD, D 99.5% purity) methanol was purchased from Sigma-Aldrich and used without further purification. The samples were enclosed in an aluminum can with a diameter of ≈ 3 cm and a height of ≈ 5 cm. An aluminum insert was used to define an annular geometry for the sample with a thickness of 0.5 mm. Measurements were performed at room temperature. Multiple scattering was deemed negligible for the coherent scattering but is probably a non-negligible contribution to the incoherent scattering and it is also going to reduce the 2/3 neutron spin flipping probability of the incoherent scattering. Multiple scattering correction were not however carried out as the main focus of the present work was on the feasibility of the experiment and on the investigation of the collective dynamics at the prepeak.

2.2. The Multi-Axis Crystal Spectrometer (MACS)

The MACS spectrometer operates a system of multiple analyzers and detectors that comprises 20 identical channels surrounding the sample. Each channel contains a vertically focusing double-crystal analyzer with two detectors. Such a design implies that one can simultaneously collect data with the given final neutron energy for the spectroscopic channel, and without energy selection for the diffraction channel. The experiment was conducted at a fixed final energy of 3.7 meV and an incident energy range from 2.95 meV to 4.45 meV centered at 3.7 meV with an energy increment of 50 μ eV. For unpolarized experiments the corresponding incident energy resolution spans from 0.13 meV to 0.29 meV. Such an instrument configuration was chosen for optimization of energy resolution and polarized neutronic performance. Data were collected for 6 h and 24 h for the non spin flip and spin flip spectra, respectively.

2.3. Wide angle polarization analysis capability on MACS

Wide-angle polarization analysis capability on MACS has been developed using ³He neutron spin filters (NSFs) and described in detail in



Fig. 1. Top view of the layout of the spin filter apparatus in the MACS sample area (top picture). Cold incident neutrons are polarized by a polarized ³He cell that is located in an RF end-compensated solenoid surrounded by an aluminum shielding box and focused at the sample position. After scattering from the sample, neutrons are spin-analyzed by a horseshoe-shaped wide angle ³He analyzer (bottom picture) and detected with a 20-detector bank system.

previous papers [23-25]. Here we outline the polarized beam setup. The polarized beam apparatus consists of a cylindrical ³He spin filter cell in a radio frequency(RF)-shielded square solenoid to polarize the incident neutron beam and a horseshoe-shaped wide angle ³He cell (shown in Fig. 1) located around the sample to spin-analyze the entire scattered beam with a wide-angle coverage of 240° and a uniform neutron path length for every scattered neutron ray [23]. Both the polarizer and analyzer cells and the sample are contained within a uniform magnetic field up to 4 mT provided by a vertical, neutron compatible solenoid. Neutron spin flipping of the incident beam is accomplished by inverting the ³He polarization of the polarizer using the adiabatic fast passage (AFP) nuclear magnetic resonance (NMR) technique. A schematic of the polarized beam configuration is shown in Fig. 1. The polarized incident flux at the sample position is 3×10^7 neutrons/(cm²s) at 3.7 meV. During the experiment, free induction decay (FID) NMR is used to monitor the ³He polarization in the spin filter cells. The ³He gas is polarized in sealed cells in an off-line lab using the spin-exchange optical pumping (SEOP) method that has recently been upgraded and expanded for polarized beam applications on MACS [26], and the cells are transported to the beam line in a portable solenoid.

3. Data reduction

3.1. Polarization efficiency correction

Since the neutron polarizing efficiencies of these ³He polarizers and analyzers are less than 100%, a correction to the raw data for the small leakage of the minority spin state is necessary before further data analysis is done. The neutron spin states of the incident and scattered beams can be either "up" or "down", hence four possible spin dependent cross sections, I^{uu} , I^{ud} , I^{du} , and I^{dd} , can be measured, where the first (second) letter, either "u"or "d", refers to the spin state of the incident and scattered beam, respectively. I^{uu} and I^{dd} are referred to as non-spinflip scattering, I^{ud} and I^{du} are referred to as spin-flip scattering. Since the ³He polarization loss was determined to be 3×10^{-4} and 6×10^{-5} per flip for the polarizer and analyzer, respectively, and the ³He polarization inversion process did not occur often, the tiny loss in ³He polarization during neutron spin flipping is neglected. The sample field was set at 2.9 mT, which was optimized for the ³He NSF performance. Since the entire polarized beam setup is contained in the solenoid that provides the field to maintain the ³He polarization, the neutron spin transport efficiency is assumed to be 100% and no neutron spin depolarization will occur. For any polarized MACS experiment in the separation of coherent from spin-incoherent scattering, it is always valid to assume that $\sigma^{uu} = \sigma^{dd}$ and $\sigma^{du} = \sigma^{ud}$, where σ^{uu} and σ^{dd} are the nonspin flip scattering cross sections from the sample, and σ^{ud} and σ^{du} are the spin flip scattering cross sections from the sample. Hence measurements of spin-dependent cross sections are reduced from four to two. In practice we measured $I^{uu} = I^{dd} = I^{NSF}$ and $I^{du} = I^{ud} = I^{SF}$. Now, we can write the two measured spin-dependent intensities in a linear combination of σ^{uu} and σ^{du} .

$$I^{\rm NSF} = (T_{\rm P}^+ T_{\rm A}^+ + T_{\rm P}^- T_{\rm A}^-)\sigma^{\rm uu} + (T_{\rm P}^+ T_{\rm A}^- + T_{\rm P}^- T_{\rm A}^+)\sigma^{\rm du} + B$$
(1)

$$I^{\rm SF} = (T_{\rm P}^+ T_{\rm A}^- + T_{\rm P}^- T_{\rm A}^+)\sigma^{\rm uu} + (T_{\rm P}^+ T_{\rm A}^+ + T_{\rm P}^- T_{\rm A}^-)\sigma^{\rm du} + B$$
(2)

where the subscript "P" or "A" refers to as either the polarizer or the analyzer. *B* is the instrument background, and was determined to be negligible. T^+ and T^- are the transmission with neutron spin parallel (+) or antiparallel (-) to the ³He spin, respectively, and are given by

$$T^{\pm} = T_{\rm E} \exp\left[-O(1 \mp P_{\rm He})\right] \tag{3}$$

where $T_{\rm E}$ is the transmission of the ³He glass cell windows, and $P_{\rm He}$ is the ³He polarization. *O* is the opacity (gas thickness) of the cell, which is linearly proportional to the neutron wavelength, ³He gas density and the neutron path length through the cell. The transmission for an unpolarized neutron beam passing through a polarized ³He cell is then given by

$$T_{\rm n} = T_{\rm E} \exp(-O) \cosh(OP_{\rm He}) = T_0 \cosh(OP_{\rm He})$$
(4)

where T_0 is the transmission for an unpolarized neutron beam passing through an unpolarized ³He cell ($P_{\text{He}} = 0$) and is given by

$$T_0 = T_{\rm E} \exp(-O) \tag{5}$$

The ³He polarization can be individually determined for each polarizer and analyzer cell from measurements of T_0 , T_n and T_E using Eqs. (4) and (5).

During the experiment, the ³He polarization decays exponentially with its own characteristic relaxation time, T_1 , for each polarizer and analyzer. Relaxation times were obtained from the initial and final ³He polarization measurements for each polarizer and analyzer cell. Relaxation times were also determined from FID (free induction decay) NMR measurements during the experiment. Relaxation times determined from the two methods agreed well. All coefficients in Eqs. (1) and (2) are determined from the initial ³He polarizations and T_1 s of both the polarizer and analyzer at any given time during the experiment. σ^{uu} and σ^{du} from the sample are then solved for any given data point.



Fig. 2. The flipping ratio measured with a pyrolytic graphite sample after installation of freshly polarized polarizer and analyzer cells. The flipping ratio is defined as the ratio of the intensity with the flipper off (filled circle) over the intensity with the flipper on (open circle) and was 19.0 \pm 0.3. The error bars are smaller than the data symbol.

After transport to the neutron beam line, the initial ³He polarizations were 84% and 74% for the ³He polarizer cell and analyzer cell, respectively. For the experiment period of 63 h, relaxation times were 251 h and 255 h for the ³He polarizer cell and analyzer cell, respectively. The flipping ratio decayed from 19 to 9, and the overall transmission as compared to the unpolarized beam configuration decayed from 18% to 9%. An example of a typical flipping ratio measurement with freshly polarized ³He polarizer and analyzer cells is shown in Fig. 2.

3.2. Determination of the coherent and incoherent contribution

The coherent and incoherent scattering spectra were determined from the non spin flip and spin flip scattering according to standard formulas considering that polarization efficiency effects have been already taken into account:

$$S_{coh}^{exp,n}(Q, E) = I^{NSF}(Q, E) - \frac{1}{2}I^{SF}(Q, E)$$

$$S_{inc}^{exp,n}(Q, E) = \frac{3}{2}I^{SF}(Q, E)$$
(6)

The coherent and incoherent scattering are directly related to the atomic motion of the system:

$$S_{coh}^{exp,n}(Q, E) \propto S_{coh}^{n}(\mathbf{Q}, E) = \frac{1}{N} \sum_{\alpha=1}^{n} \sum_{\beta=1}^{n} b_{\alpha}^{coh} b_{\beta}^{coh} \sqrt{N_{\alpha} N_{\beta}} S^{\alpha\beta}(\mathbf{Q}, E)$$
(7)

$$S_{inc}^{exp,n}(Q,E) \propto S_{inc}(Q,E) = \sum_{i_{\alpha}=1}^{N_{\alpha}} \frac{1}{N_{\alpha}} (b_{\alpha}^{inc})^2 S_{self}^{\alpha}(Q,E)$$
(8)

where *N* is the total number of atoms, b_{α}^{coh} and b_{α}^{inc} are the coherent and incoherent scattering lengths of the α atoms, respectively, and the dynamic structure factors are defined as Fourier transforms of the corresponding Intermediate Scattering Functions (ISFs):

$$S^{\alpha\beta}(\mathbf{Q}, E) = \frac{1}{2\pi\sqrt{N_{\alpha}N_{\beta}}} \sum_{i_{\alpha}=1}^{N_{\alpha}} \sum_{i_{\beta}=1}^{N_{\beta}} \int_{-\infty}^{\infty} I^{\alpha\beta}(Q, t) e^{\left(-i\frac{E}{\hbar}t\right)} dt,$$
(9)

$$I^{\alpha\beta}(Q,t) = \langle e^{\{i[\mathbf{Q}\cdot\mathbf{R}_{i\alpha}(t) - \mathbf{Q}\cdot\mathbf{R}_{i\beta}(0)]\}} \rangle$$
(10)

$$S_{self}^{\alpha}(Q, E) = \frac{1}{2\pi N_{\alpha}} \sum_{i=1}^{N_{\alpha}} \int_{-\infty}^{\infty} I_{self}^{\alpha}(Q, t) e^{\left(-i\frac{E}{\hbar}i\right)} dt$$
(11)

$$I_{self}^{\alpha}(Q, t) = \langle e^{\{i[\mathbf{Q}\cdot\mathbf{R}_{i\alpha}(t)-\mathbf{Q}\cdot\mathbf{R}_{i\alpha}(0)]\}} \rangle$$
(12)

 N_{α} being the number of atoms of the α specie in the sample, $\mathbf{R}_{i_{\alpha}}$ the position of the i-th α atom, and $\langle ... \rangle$ indicates an ensemble average.

4. Data analysis

The experimental spectra were analyzed using the sum of a Lorentzian function and an energy independent background:

$$S(Q, E) = \frac{A(Q)}{\pi} \frac{\Gamma(Q)/2}{E^2 + [\Gamma(Q)/2]^2} \otimes R(Q, E) + BKG_0(Q)$$
(13)

where Γ is the Full Width at Half Maximum (FWHM) of the Lorentzian, A(Q) represents the intensity of the Lorentzian, and R(Q,E) is the instrumental resolution. The fitting parameter BKG₀ accounts for QENS components much broader than the instrumental energy window; in the present case, such broad feature originates from both the ³He cell glass [27] as well as from methanol, which is known to have a sub-picosecond relaxation component [6]. Because of the presence of this very broad component, whose area cannot be determined within the limited energy window, the total spectral intensity, i.e. the structure factor, or the relative weight of the slow and fast relaxation components cannot be determined. The above fitting scheme assumes simple exponential relaxation processes with a relaxation time proportional to the inverse of Γ . R(Q,E) was modeled to be a Gaussian function with a Full Width at Half Maximum (FWHM) determined from the results of the fitting of data taken on an aluminum sample under the same experimental condition of the sample. The resolution FWHM values range from ≈ 0.24 meV at low Q to ≈ 0.20 meV in the intermediate and high Q range probed. The use of an aluminum sample for determining the resolution function is probably not ideal; vanadium, a purely incoherent scatterer, is employed most commonly; given that the interest was focused on the coherent scattering the use of a null matrix of $Ti_{2.08}$ Zr [28] would probably had been a good option, which however was not available. The aluminum sample which has no dynamics in the window of relevance is mostly a coherent scatterer but was neverthless counted sufficiently long to obtain the required energy resolution with satisfactory precision for both the NSF and SF measurements. The fitting of the data was carried out using the utility PAN in DAVE [29]. The spectra at $Q = 1.1 \text{ Å}^{-1}$, together with the fit results and components are shown as examples in Fig. 3.

5. Results

Fig. 4 reports the main results of the investigation. In plot a) the Q dependence of the total spectral area, A, as obtained from the fits of $S_{coh}^{exp,n}(Q, E)$ is shown; a maximum in correspondence of the prepeak is observed confirming that the spectra is associated with the coherent signal.

Fig. 4 b) reports the Q dependence of the Γ for both the coherent and incoherent spectra. Up to a Q value of $\approx 0.85 \text{ Å}^{-1}$ the incoherent data follow a Fickian diffusive behavior: $\Gamma = 2DQ^2$, D being the self diffusion coefficient of methanol. The obtained diffusion coefficient is $(3.43 \pm 0.02) \times 10^{-9} \text{ m}^2/\text{s}$. This value is in reasonable agreement with previous results from Pulsed Gradient Spin Echo (PGSE) nuclear magnetic resonance on CH₃OD [30], which report the experimental value of $2.08 \times 10^{-9} \text{ m}^2/\text{s}$ at 294 K and would estimate a value of $2.81 \times 10^{-9} \,\text{m}^2/\text{s}$ at 298 K on the basis of a parametrization using the empirical Vogel-Tammann-Fulcher (VTF) equation. The difference between the present and the PGSE results might be, at least partly, explained by multiple scattering which will tend to broaden the spectra. At higher Q values, other dynamic processes such as molecular rotation and the rotation of the methyl groups are expected to become increasingly relevant. A more complex model would be required which is not warranted for the present results and would be beyond the scope of the present work. The Q dependence of Γ for the coherent component deviates significantly from a diffusive behavior. At the lowest Q studied here, the value of the coherent and incoherent dynamics are similar, in agreement with a previous report from MD simulations [6]. At higher Q values the collective dynamics probed by the coherent signal is significantly slower than the single particle dynamics probed by the



Fig. 3. Fit of the experimental data according to Eq. (13) at $Q = 1.1 \text{ Å}^{-1}$. The symbols are experimental data, the continuous black line is the fit; the red dashed, blue dotted, and green dash-dotted lines represent the Lorentzian function before convolution, the linear background, and the height scaled resolution function respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

incoherent contribution. This finding can be explained by the de Gennes' narrowing, $\Gamma \sim 1/S(Q)$, a slowing of the dynamics at Q values corresponding to peaks in the structure factor, i.e corresponding to characteristic length scales of the system [31]. Also this result is in agreement with recent simulation and neutron scattering investigation. The characteristic relaxation time $\tau = 2\hbar/\Gamma$ is (4.7 ± 0.2) ps, consistent with values reported previously for the dynamics of the molecular associates [7]. It should be kept in mind, however, that previous results referred to the time scale of the relative dynamics of the hydroxyl groups with respect to the methyl groups [7]; here, in the coherent spectra, the collective dynamics of all the partial dynamic structure factors, weighted as detailed in Eq. (7), is probed.

6. Conclusions

A new prototype measurement capability for separating coherent quasi-elastic neutron scattering spectra from the spin incoherent spectra has been demonstrated for a partially deuterated (CH₃OD, D 99.5% purity) methanol sample. We have developed an adequate procedure for taking data, performing polarization efficiency corrections and further data analysis for the separation of coherent and spin incoherent scattering for the wide angle polarization analysis capability on MACS. Collective dynamics of the prepeak in partially deuterated methanol determined from coherent scattering agrees with those from Refs. [6,7] and the incoherent data yield a self diffusion coefficient value in agreement with previous results [30]. These results show that the MACS wide-angle polarization analysis capability can be applied to



Fig. 4. a) The area of the coherent spectra. Notice the location of the maximum in correspondence of the prepeak. b) The FWHM values obtained from the fitting of the coherent and incoherent spectra using Eq. (13). The dash-dot red line represents the fit of the incoherent spectral width to a Fickian diffusion model in the range up to Q = 0.85 Å^{-1} . Notice how the results of the coherent signal do not follow the Q² behavior of the Fickian diffusion and a slowing down of the dynamics can be observed in correspondence of the prepeak. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

both hard condensed matter and soft matter studies.

After this successful feasibility test of separation of coherent and spin incoherent scattering for investigations of collective and spin particle dynamics, we proceed to discuss future possibilities. During the feasibility test the sample was limited to the room temperature. We have integrated a closed cycle refrigerator (CCR) with a temperature range from 3 K to 800 K that allows for a sample size of 60 mm. The tail section of the CCR where the sample is located is still small enough to be inserted inside the horseshoe-shaped ³He analyzer cell developed recently [23]. Since the CCR is in close proximity to both ³He polarizer and analyzer, we have measured the relaxation time of the ³He polarization with and without the CCR and found there is no effect to the relaxation time when the CCR is installed. These implemented improvements would further expand the capability allowing polarized beam experiments for soft matters done at most common sample environment conditions.

In conclusion, the use of opportunely shaped ³He cells offers new opportunities for the separation of the coherent and spin incoherent nuclear scattering over a wide scattering angle range, although it introduces the complexity of a time dependent polarization efficiency and the necessity of periodically exchanging the neutron polarizing/analyzing devices. The significant penalty in counting rates, inherent with the use of polarization devices, could be overcome by the use of an instrument, such as MACS, with a very high incoming flux, due to a

double focusing monochromator, and equipped with an array of energy analyzer crystals for efficient neutron detection. The energy resolution which can be achieved on MACS is comparable to that of other instruments currently offering similar polarization analysis capabilities [10,11], but it might represent the main limiting factor for the use on MACS for soft matter application. The deployment of the horseshoe shaped wide angle ³He analyzer, in conjunction with a suitable polarizing guide element, on a time of flight machine, such as the Disk Chopper Spectrometer (DCS) at NIST [32], would allow to perform measurements with much better energy resolution; however, the much reduced flux at DCS compared to MACS. ≈ 1000 times smaller. represent a serious challenge. Even so, such efforts have been undertaken. with promising initial results, on the cold neutron multichopper spectrometer LET [17]. Nevertheless, we envisage that new insight in the dynamics of systems such as, for example, molecular [7] and ionic [10] liquids as well as polymers [11] can be obtained with the use on MACS of the neutron polarization analysis capability with a large momentum transfer coverage.

Disclaimers

Identification of a commercial product does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the product is necessarily the best for the stated purpose.

Throughout the paper error bars and uncertainties represent one standard deviation.

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