Nuclear Instruments and Methods in Physics Research A 754 (2014) 10–14

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

# Proton polarization above 70% by DNP using photo-excited triplet states, a first step towards a broadband neutron spin filter

T.R. Eichhorn [a,b], N. Niketic [a], B. van den Brandt [a], U. Filges [a], T. Panzner [a], E. Rantsiou [a], W.Th. Wenckebach [a], P. Hautle [a,*]

[a] Laboratory for Developments and Methods (LDM), Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland
[b] Laboratory of Functional and Metabolic Imaging, École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland

ARTICLE INFO

*Article history:*
Received 18 February 2014
Received in revised form
21 March 2014
Accepted 21 March 2014
Available online 29 March 2014

*Keywords:*
Neutron polarization analysis
Spin filters
Dynamic nuclear polarization
Photo-excited triplet states
Neutron optics

ABSTRACT

The use of polarized protons as neutron spin filter is an attractive alternative to the well established neutron polarization techniques, as the large, spin-dependent neutron scattering cross-section for protons is useful up to the sub-MeV region. Employing optically excited triplet states for the dynamic nuclear polarization (DNP) of the protons relieves the stringent requirements of classical DNP schemes, i.e low temperatures and strong magnetic fields, making technically simpler systems with open geometries possible.

Using triplet DNP a record polarization of 71% has been achieved in a pentacene doped naphthalene single crystal at a field of 0.36 T using a simple helium flow cryostat for cooling. Furthermore, by placing the polarized crystal in a neutron optics focus and de-focus scheme, the actual sample cross-section could be increased by a factor 35 corresponding to an effective spin filter cross-section of $18 \times 18$ mm$^2$.


## 1. Introduction

Dynamic nuclear polarization (DNP) [1] is traditionally used to create polarized targets to investigate the role of spin in nuclear and particle interactions. But it has also opened new possibilities in neutron science where the strong spin dependence of the neutron scattering on protons is exploited [2–4]. Proton spin polarizations near unity have been achieved employing the classical DNP scheme: electron spins are polarized by cooling them down to low temperature (~1 K) in a strong magnetic field (2.5–5 T) and their high polarization is transferred to the nuclear spins by means of a microwave field. For some applications the necessary cryogenics and magnets are prohibitive. A solution is the more recent method of "triplet DNP" that uses the electron spin of optically excited triplet states [5]. The triplet electron spin is polarized as a result of the selection rules of the optical excitation process. Neither low temperatures nor high fields are required and the restrictions for applications are relieved. Recently we have demonstrated that triplet DNP can be used to build a reliably working spin filter for neutrons operating at 0.3 T at a temperature of 100 K [6], and that a sizeable proton spin polarization can be achieved in single crystals of naphthalene doped with pentacene [7].

In this note we present results which we believe to be a substantial step towards the application of triplet DNP to a neutron spin filter: we achieved a proton spin polarization above 70% and integrated the filter in a neutron focusing arrangement.

## 2. Neutron spin filtering with polarized protons

Shapiro and coworkers demonstrated that dynamically polarized proton spins offer an attractive possibility to realize a broad band neutron spin filter, as the spin-dependent neutron–proton cross-section is large in a broad wavelength range [8]. But so far an actual implementation of a polarized proton spin filter has been restricted to a few special cases [9–12]. This is most probably due to the necessary cryogenics and magnets needed for a classical DNP system.

The working principle of a polarized proton spin filter is based on the fact that the singlet cross-section for neutron–proton scattering is much higher than the triplet cross-section [24]. Hence, neutrons which are polarized anti-parallel to the protons are much stronger scattered than those polarized parallel. This applies for coherent and incoherent scattering. It is customary to write the cross-section as

$$\sigma_{\pm} = \sigma_0 \pm \sigma_p P \quad (1)$$

where the + sign and the − sign stand for the two eigenstates of the neutron spin with respect to the direction of the proton spin

* Corresponding author. Tel.: + 41 56 310 3210.
*E-mail address:* patrick.hautle@psi.ch (P. Hautle).

http://dx.doi.org/10.1016/j.nima.2014.03.047
0168-9002/

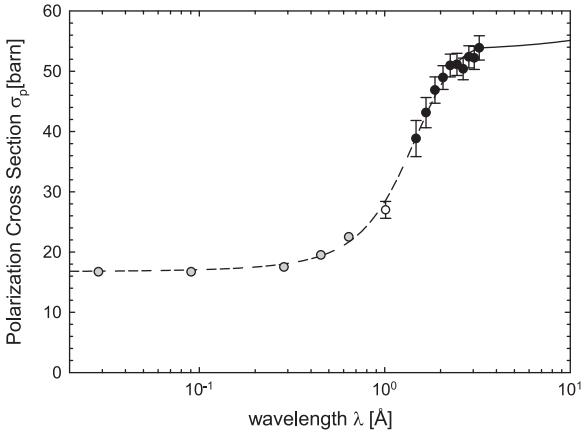


**Fig. 1.** Typical trend of the polarization cross-section $\sigma_p$ as function of the neutron wavelength. The data points are a compilation of literature values: black dots and solid line [6], open dot [11], gray dots [8]. The dashed line is to guide the eye.

polarization. The second term is the product of the "polarization cross-section" $\sigma_p$ and the proton spin polarization $P$. The first term $\sigma_0$ is either independent of $P$ or a symmetric function of $P$.

After passing through a spin filter with a proton density $N$ and a thickness $d$, the two spin components of the neutron beam are attenuated by a factor $\exp(-\sigma_{\pm} Nd)$. Thus, after passing through the filter an initially unpolarized neutron beam has a polarization

$$A = \tanh(\sigma_p PNd) \tag{2}$$

which is called the filter analyzing power. The thicker the filter, the higher the filter analyzing power for given $(\sigma_p N)$, and the lower the transmission

$$T = \exp(-\sigma_0 Nd)\cosh(\sigma_p PNd) \tag{3}$$

of the neutron beam. Note that for a given thickness $d$, the total transmission $T$ increases with the filter polarization $P$.

Structural effects on the values of $\sigma_p$ and $\sigma_0$ observed for slow neutrons are difficult to predict theoretically even when the crystal structure is well known. So these cross-sections need to be determined experimentally for each material. Fig. 1 gives the trend for the value of $\sigma_p$ as a function of the incoming neutron wavelength. Here the black dots and solid line are the experimental values for naphthalene [6].

## 3. Experimental methods

A proven system for triplet DNP is a naphthalene crystal doped with a small concentration of pentacene guest molecules. Pentacene is optically excited into a short lived triplet state with a strongly aligned electron spin $S=1$. In an external magnetic field two electron spin resonance (ESR) lines are observed at $\sim$0.3 T corresponding to the transitions $S_z = -1 \leftrightarrow 0$ and $S_z = 0 \leftrightarrow +1$. For $B \| X$ the ESR lines are separated by about 1.5 GHz. Also the $S_z=0$ level is strongly favored. In triplet DNP only one of the two ESR transitions is used and the two energy levels involved may be regarded as an effective spin-$\frac{1}{2}$ system with an "effective" electron spin polarization $P \sim 0.9$ [13].

This high electron polarization is most efficiently transferred to the nuclear spins by means of the Integrated Solid Effect (ISE) [14,15]. In this method the externally applied magnetic field is adiabatically swept over the inhomogeneously broadened ESR line (typically in 10 μs) under microwave irradiation, which allows all electron spins to contribute constructively to the polarization transfer. Provided the proton spin–lattice relaxation is long enough, the cycle can be repeated many times and the proton spin polarization is accumulated and can approach the electron spin polarization. Since the proton spin–lattice relaxation time in naphthalene is already very long at liquid nitrogen temperature ($\sim$24 h in a field of 0.3 T), triplet DNP experiments need moderate cryogenic means only.

The sample is cut out of a large single crystal grown with a self-seeding vertical Bridgman technique from zone-refined naphthalene doped with pentacene-$d_{14}$. The pentacene concentration is determined with optical transmission spectroscopy to be $(3 \pm 0.5) \times 10^{-5}$ mol/mol. The size of the filter was $4.3 \times 4.8 \times 5.1$ mm$^3$, where its bottom/top corresponds to the crystal *ac*-plane. It is mounted on a polychlorotrifluorethylene (PCTFE) holder, introduced in a helium flow cryostat and cooled to temperatures between 25 K and 100 K. The pentacene molecules are excited into the triplet state with a disk laser system operating at 515 nm generating pulses with a width of 400 ns at a repetition rate between 400 Hz and 4 kHz. The light is transported via a multimode fiber to an optical stage at the bottom of the cryostat which collimates the unpolarized light to a beam waist of about 9 mm. The beam axis is vertical and along the *b*-axis of the sample crystal, which is illuminated homogeneously. The energy per pulse at the entrance window of the cryostat is approximately 1 mJ. A small electromagnet provides a static magnetic field of about 0.36 T and DNP was performed on the so-called high field transition ($S_z = 0 \leftrightarrow 1$) using a pulsed ESR system operating at 9.3 GHz and synchronized to the laser. The latter also enables us to observe the ESR signal of the triplet states and to orient the static magnetic field along the *X*-axis of the pentacene molecules. A pulsed NMR system is used to monitor the proton polarization and to optimize the parameters for the ISE process. A detailed description of the triplet DNP apparatus is given in [16].

The neutron spin filter experiment was performed at the new BOA beam line, an upgrade of the former FUNSPIN beam [17], at the continuous spallation neutron source SINQ at the Paul Scherrer Institute in Switzerland. A layout of the experimental set up is given in Fig. 2. The incoming beam is polarized by the original bender and its spectral distribution is determined by the liquid deuterium moderator. The inherently large beam divergence is restricted by two collimators [18] to vertically 0.28° and horizontally 0.16°. With an adiabatic spin flipper the neutron polarization can be reversed with respect to the polarity of the guide field at the polarized target. The neutron spin polarization is maintained throughout the beam path to the sample with guide fields using permanent magnets. The triplet apparatus is placed between two parabolic supermirror lenses. The lenses form a focus–defocus pair for the neutron beam, both in the vertical and in the horizontal planes. The focusing lens in front of the filter is 1.0 m long and accepts a beam area of 25 × 25 mm$^2$ and has an exit size of 13.363 × 13.363 mm$^2$. A defocusing lens of identical focal length is located after the spin filter. It is only 0.5 m long with a correspondingly smaller exit size of 20.4 × 20.4 mm$^2$ and restores the original beam properties in the far field. Neutron ray-trace simulations with McStas [19] show that for the present beam collimation, the transmission of the lens system extends to below 1 Å and is constant from about 2 Å upwards, thus the lenses marginally modify the incoming cold spectrum that is peaked at 3 Å. A detailed description and characterization of the lenses is

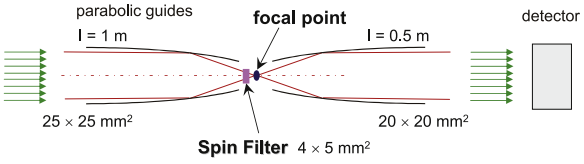


**Fig. 2.** Schematic representation of the neutron lenses setup.