A Dynamic Nuclear Polarisation system based on a continuous-flow 4He cryostat

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Introduction

In the field of MRS/MRI, "hyperpolarisation" refers to a liquid solution in which certain nuclei have a polarisation considerably above the equilibrium value at physiological temperature and practical magnetic field strength. Dynamic Nuclear Polarisation (DNP) [1] is the basic step in one among several approaches to hyperpolarisation. The usual DNP sample is solid, typically kept at temperatures around 1 K, such as the polarised targets used in particle physics [2]. The extension of the DNP technique to hyperpolarisation was made recently, when it was shown that such a cold solid can be transformed into a room temperature solution, while preserving a high polarisation [3]. Here we describe a DNP system that presents several interesting novelties. Its performance is discussed on the example of ^{13}C in glycine, but it has also been used to polarise other nuclei in frozen glassy solutions, such as ^{1}H, ^{2}H and ^{6}Li.

System Description

The sample cryostat has been specifically designed such that its tail fits inside the standard 88 mm RT bore of a low-field NMR magnet (the experiment runs at 143 MHz protons). It takes He from a storage dewar close to the magnet through a U-shaped transfer line. This transfer line is a rigid single piece, and is kept as short as possible. It is not vapor-cooled, rather the enthalpy of the cold up-streaming gas is used inside the cryostat. From the transfer line the He enters into a carefully optimised gas/liquid separator, from which the liquid reaches the sample space (37 mm diameter) through an adjustable impedance. The sample is thus maintained under superfluid helium, pumped to below 1.1 K by a 250 m^3/h Rootes pump. Sample preparation was according to procedures used for polarised targets [4]. The solvent is a glass-forming mixture of water with ethanol or glycerol, and the sample is in the form of 2 mm frozen droplets, to improve the cooling efficiency. As paramagnetic center the nitroxyl radical TEMPO was used, which was found to dissolve readily in the solvents used. The microwave cavity is a cylinder of 28 mm inner diameter and 38 mm height, open at the top. It is coupled to a 94 Ghz microwave source, operated at 30 – 60 mW power level, through a rectangular waveguide, a transition to cylindrical guide and a tapered horn. Inside the cavity is a removable Teflon sample holder of 0.6 ml volume. For the solid-to-solution transformation, the coupling structure was pulled out, and replaced by a dissolution mechanism which was developed similar to recent approaches described in [3]: steam (5 ml water, heated to 170 C, and propelled by He gas at 6 bar) is sent down through a capillary, and dissolves the solid. Within 5 sec, the gas stream drives the resulting solution out of the polariser magnet through a second, 6 m long, capillary to the isocenter of the actively-shielded 9.4 T animal scanner (Varian/Magnex), where the sample is collected in a hydraulically driven all-Delrin injection pump, that separates the solution from the gas, and injects a chosen amount into an animal or a phantom.

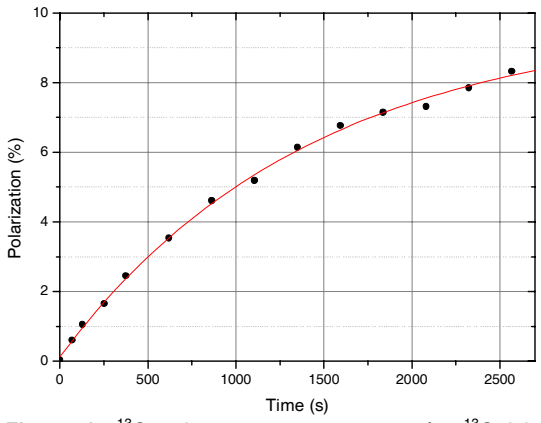

Figure 1. ^{13}C polarisation versus time of 1-^{13}C labeled glycine obtained with a microwave power of 30mW. The polarisation time constant is about 1500 seconds.

Performance

The liquid He consumption during a typical working week was less than 60 l. A satisfactory ^{13}C polarisation, around 8% in glycine, was obtained with 30-45 min of irradiation at 30 mW. Introducing a new sample and cooling to 1.1 K requires 20-30 min, so that the system is capable of producing polarised liquid samples approximately every hour. We obtain fast polarisations and good final values using low microwave power levels, although we use the "simple" radical TEMPO (first synthesised fifty years ago), instead of a triarylmethyl (TAM) radical designed for the purpose [3]. Three factors of technical and theoretical nature, that potentially contribute to this efficiency, were identified: First, a suitable geometry of the microwave coupling structure improves the mode pattern in the cavity, and thus the polarisation rate. Second, the ESR lineshape of TEMPO is determined by g-anisotropy, while the TAM radicals have small spin-orbit coupling. Smaller spin-orbit couplings usually lead to narrower lines but longer spin-lattice relaxation times. Schematically, narrow lines are favourable, but long relaxation times are not. Third, at least theoretically [1] it is possible to obtain a very high polarisation also with radicals that have g-anisotropy, by going to low enough temperatures. We conclude that a high turnover of highly polarised liquid samples can be obtained with a system based on a high-performance cryostat that fits into a standard vertical bore NMR magnet, and equipped with optimised microwave hardware, while using a standard radical as paramagnetic center.

Acknowledgements

This work is supported by SNSF grant 200021-109479, and by the Fund of the Dean of the Faculty of Basic Sciences, EPFL. Supported by the Centre d'Imagerie BioMedicale of the UNIL, EPFL, UNIGE, CHUV and HUG, and by the Jeantet and Leenaards Foundations.

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