Nuclear hyperpolarization in 1H and 19F rich fluids induced by photon beams endowed with Orbital Angular Momentum

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INTRODUCTION We report a novel method for generating high levels of nuclear magnetic polarization within a volume of fluid using tightly focused beams of photons that are endowed with Orbital Angular Momentum (OAM). We measured nuclear magnetic polarization in excess of 1.5%, at room temperature, with samples containing 1H and 19F that were irradiated with only up to 5s of continuous wave (CW) visible wavelength laser beams endowed with an OAM charge of up to 20 and focused to approximately 10μm beam waist. It is important to mention that, unlike other hyperpolarization methods (e.g. DNP, spin exchange optical pumping, PHIP), this novel nuclear hyperpolarization method produces a localized and quickly forming (of the order of seconds) magnetic polarization in fluids at room temperature and potentially also in gases, solids and living tissue.

THEORETICAL FRAMEWORK Photon–molecule interactions obey basic energy and momenta conservation laws. For example, the hyperpolarization technique known as “spin exchange optical pumping” takes advantage of the conservation of the spin during a photon–electron collision. The nuclear Overhauser effect extends the spin momentum conservation to large populations of electrons and nuclei. For the DNP case, the nuclear momenta are oriented towards a privileged spatial direction through a chain of fine and hyperfine interactions that could be viewed as spin and angular momentum conservation rules involving the molecular orbital momenta, electron and nuclear spins. Our vectorial hyperpolarization technique takes advantage of the vectorial transfer of the photon OAM to the molecular momenta, and subsequent re-orientation of the nuclear momenta, through aforementioned fine and hyperfine interactions, towards a privileged direction of the photon OAM momentum (for his work, same as the direction of propagation of the photon beam). It has been shown theoretically[1] and experimentally[2] that photons are able to carry OAM in addition to linear and spin momenta. The OAM (also known as optical vortex) is quantized, within the QED formalism, in integer multiples of the reduced Planck constant. One of the first examples of a rotational momentum transfer from light vortices to atoms is depicted in the work of Babiker et al.[3], which shows that the amplitude of the torque imparted to an absorbing atom is proportional to the OAM charge. Alexandrescu et al.[4] formulated the transition probabilities of the molecular rotational and electronic motions as a function of the OAM charge, initiated a qualitative discussion on the associated transition selection rules and concluded that the OAM of a photon is transferred to the molecular rotational and vibrational momenta.

In our attempt to demonstrate the possibility of light vortex interaction with nuclei, we first calculated the steady state probability of the interaction of the photons OAM with molecular fermion components and found that this is proportional to the square of the OAM charge and inversely proportional to the waist of the incident beam. According to this model, a fermion angular momentum vector will align with the orientation of the OAM of the incident photons, while their projection is modified within the selection rules’ boundaries by the OAM charge. We then combined the steady–state Hamiltonian of the fermion–photon OAM interaction with the time–dependent model devised by Zare[5] and the dynamics of spin diffusion model developed by Happer et al.[6], and derived a nuclear hyperpolarization coefficient expression $H_{\alpha}(\gamma) = \frac{1}{(\gamma B)(\sqrt{2}\alpha)} \left[ 1 - \exp(-\gamma t_{OAM}) \right]$, which is a simplified quantitative description of the nuclear polarization evolution for matter irradiated with photons endowed with OAM (I: beam intensity, J: OAM charge, α: molecule size, B: beam waist, γ: photon wavelength, $t_{OAM}$: hyperpolarization time constant).

MATERIALS AND METHODS This novel nuclear magnetic hyperpolarization technique was experimentally verified by irradiating fluid samples containing 1H and 19F nuclei with a 532nm CW laser beam endowed with OAM levels of 0 to ±20 and focused (using a Mitutoyo M plan Apo 100X, 0.7NA microscope objective MOBJ) to an ~10μm beam waist. One version of our experimental setup is schematically depicted in Figure 1. The OAM was imparted to the beam using computer-generated phase holograms implemented on a Holoeye PLUTO HDTV resolution phase only spatial light modulator (SLM). The OAM beam had a measured power of ~40mW and was focused inside a ~1nl fluid sample (confined within a cuvette with a cross-section of 20μm x20μm). A custom Helmholtz transmit/receive micro-coil (HC) was constructed and placed directly around the sample/photonic focal point. The setup was placed inside the bore of a 1.0T Philips Panorama MRI scanner, with the OAM beam aligned parallel to B0. Following sample irradiation under various conditions (light off, OAM=0...±20) for 1 second, a standard non-selective spin-echo measurement was made; 1000 measurements using each experimental condition were performed and averaged together. The measured NMR signal level under “light off” and OAM=0 conditions were comparable (+1 std around the average value measured under OAM=0 conditions fell within ±1 std around the average value measured using “light off”) and the measured NMR signal increased with an absolute value of the OAM (Figure 2). Taking into account the difference in sample volume inside the measurement coils (~1nl) and inside the OAM beam (between as 0.1pl and 0.4nl depending on how the photon beam’s geometry is modeled), this measured increase in NMR signal corresponds to nuclear magnetic polarization levels of between ±1.5% to 5% inside the OAM beam’s focal spot. Similar nuclear magnetic polarization levels were obtained from experiments performed with photon beams endowed with OAM of different wavelengths (646nm, 488nm, 0.1nm).

CONCLUSIONS The novel method proposed here enables nuclear hyperpolarization of liquids at room temperature using a photon beam endowed with OAM. The experimental results presented above show the feasibility of this approach using visible light beam (532nm). This approach has a potential to support applications in which biological tissue is directly hyperpolarized while inside an MR scanner; the use of higher energy photons (X-rays) may also enable subcutaneous structures to be non-invasively hyperpolarized.

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