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Development of polarized sources based on molecular photodissociation

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Molecular photodissociation is an innovative method for the preparation of polarized atoms and molecules. It is a fundamental chemical process that involves the absorption of one or more polarized photons by a molecule including its fragmentation into polarized atomic (or molecular) fragments. Recently, T. P. Rakitzis' group produced high densities of spin-polarized hydrogen atoms applying molecular photodissociation to hydrogen halides. The obtained densities (10^{19} cm^{-3}) and short production times (ns timescales) surpass by several orders of magnitude conventional methods such as spin-exchange optical pumping and Stern-Gerlach spin separation. These density and time regimes make it an ideal candidate for a broad range of applications, e.g., laser-induced acceleration from polarized gas targets and polarized five-nucleon fusion reactions (d-^{3}H, d-^{3}He). The second has been shown to have an increased cross section by ~50% compared to the unpolarized case. The photodissociation method has been adopted by M. Büscher's group for the production of polarized proton and deuteron beams at the Forschungszentrum Jülich. Here, we report on the production and detection scheme of these beams.

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1. Introduction

The development of nuclear spin polarization is of vital importance for applications in several fields, such as accelerators and polarized nuclear fusion. For instance, nuclear-spin-polarized gas targets are produced for the measurement of spin-dependent effects in conventional accelerators. Often, the target density is not adequate for the needs of some experiments and storage cells are required in order to enhance the target thickness [1]. Laser-induced acceleration is an alternative method to accelerate particle beams with an energy of several MeV or even GeV over a distance of a few cm. However, the study of the influence of electromagnetic laser and plasma fields on the beam polarization demands high fluxes [2]. The conventional methods for the production of spin-polarized hydrogen isotopes cannot produce directly the required densities for the aforementioned applications. Their limitations are discussed in the next section along with the advancements provided by molecular photodissociation.

Nuclear fusion offers an almost inexhaustible supply of zero-carbon energy. Although the research into fusion began in the 1940s, sustainability still has not been attained. The use of "polarized fuel" results in improvements that can bring us closer to this goal. Of particular scientific interest are reactions between deuterium nuclei (spin 1) alone or between deuterons and tritium or ^{3}He nuclei (spin $1/2$). The d-d reactions (four-nucleon reactions) divide into two branches with roughly equal probabilities:

$$\mathrm{d} + \mathrm{d} \longrightarrow \mathrm{n} + {}^3\mathrm{He} + 3.27\,\mathrm{MeV}, \tag{1}$$

$$\mathrm{d} + \mathrm{d} \longrightarrow \mathrm{p} + {}^3\mathrm{H} + 4.03\,\mathrm{MeV} \tag{2}$$

called the "neutron branch" and the "proton branch", respectively. The energy release appears in the form of kinetic energy of the fusion products. Experimental fusion reactors do not generally use four-nucleon, but five-nucleon reactions instead. These are given below:

$$\mathrm{d} + {}^3\mathrm{H} \longrightarrow \mathrm{n} + {}^4\mathrm{He} + 17.58\,\mathrm{MeV}, \tag{3}$$

$$\mathrm{d} + {}^3\mathrm{He} \longrightarrow \mathrm{p} + {}^4\mathrm{He} + 18.34\,\mathrm{MeV}. \tag{4}$$

The first (d-^{3}H) leads to the formation of a helium-4 (ordinary helium isotope) nucleus, a neutron, and an energy release of 17.58 MeV. The high reaction rate and the large energy release make this reaction the most favorable for controlled fusion. On the other hand, the reaction rate of d-^{3}He is significantly lower compared to d-^{3}H [3], even though the energy release is of the same order of magnitude.

The effect of polarized deuterons in the d-d reactions is poorly understood, since there is a number of problems in the computations leading to predictions that range from suppression to enhancement of the reaction rates in the energy range of 10 to 100 keV. This discrepancy can be solved by direct measurements [3, 4]. On the contrary, for the five-nucleon reactions it is known that the reactants' polarization increases the cross sections by 50% [3, 5]. It also allows the control over the emission direction of the products leading to more effective designs for the reactor walls.

2. Production of spin-polarized hydrogen isotopes

The most widely used conventional methods for the production of nuclear-spin-polarized hydrogen isotopes are the atomic beam source (ABS, based on the Stern-Gerlach experiment and

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RF transition units) and the spin-exchange optical pumping. The first has a limited beam density of the order of 10^{12} cm^{-3} yielding a production rate of $\sim 10^{17}$ H/s. The latter can achieve densities of 10^{14} cm^{-3} and fluxes of $\sim 10^{18}$ H/s but with lower polarization values compared to the first.

Molecular photodissociation is an alternative method for the production of electron- and nuclear-spin-polarized hydrogen isotopes. In general, polarized atoms can be produced from the photodissociation of diatomic molecules with circularly-polarized light. For instance, consider a diatomic molecule AB with total projection Ω of the molecular electronic angular momentum along the bond axis in the dissociative excited state, which is excited by the circularly polarized photodissociation light beam. If m_A and m_B are the projections of the atomic angular momenta J_A and J_B of the separated atoms, then Ω is adiabatically correlated to these projections:

$$\Omega = m_A + m_B. \tag{5}$$

For example, if $\Omega = 1$, and $J_A = J_B = 1/2$, then m_A and m_B are both constrained to be $1/2$ by Eq. (5), and therefore J_A and J_B will be fully polarized.

Several studies have been focused on the hydrogen halides, HY, which are the most effective molecules for the production of spin-polarized hydrogen isotopes [6]. Depending on the halogen Y and the wavelength of the UV beam (for right or left circular polarization), the photodissociation mechanism is determined and the subsequent spin projection of H is defined [7, 8]. For instance, the photodissociation of DI with right-circularly polarized light at 266 nm yields D atoms with electron spin down, whereas at 213 nm the spin projection is reversed.

The photodissociation method is more efficient compared to conventional methods, since every absorbed UV photon can produce one spin-polarized hydrogen isotope with up to 100% electron polarization. This can be transferred to the nucleus within half of the hyperfine period in the absence of external fields via the hyperfine quantum beats. Thus, the production timescales are also several orders of magnitude faster. These advantages led to the observation of 10^{19} cm^{-3} of spin-polarized deuterium [9] and 10^{20} cm^{-3} of spin-polarized hydrogen [10]. The experiment and the depolarization mechanisms are described in the next section.

3. High-density spin-polarized hydrogen isotopes observed via magnetization quantum beats

Polarized samples produced from molecular photodissociation with short UV light pulses posses a large time-dependent magnetization. This can be detected with a pickup coil similar to NMR. Such an experiment was developed by T. P. Rakitzis' group in 2018 [9]. The main parts of the experimental setup are: a photoelastic modulator, a quarter-wave plate, a focusing lens, and a pickup coil placed in a small vacuum chamber (containing the hydrogen halide gas). The circular polarization of the 150 ps, 213 nm or 266 nm, light pulse (with energy 1−10 mJ) is alternated between right and left on a shot-to-shot basis at a 10 Hz repetition rate by the first two components. Then, the laser is focused through the pickup coil and photodissociates hydrogen halide gas at pressures 0.1−5 bar. The total number of absorbed photons depends on the sample thickness and the focusing conditions. Essentially, it is equal to the number of spin-polarized hydrogen isotopes produced from photodissociation. The electrons of the H isotopes are initially polarized, whereas the nuclei are unpolarized. Consequently, the electron polarization is transferred to the nuclei

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and back at the characteristic hyperfine frequency of the atom. This results in a time-dependent electron magnetization which is directly detected by a pickup coil. The alternating polarization of the light leads to signal reversal and the subtraction of individual signals from two consecutive pulses suppresses the background noise from the operation of the UV laser. Further details about the experimental setup and measurements can be found elsewhere [7–10].

Two density regimes are observed depending on the focusing geometry [11]. If the laser beam is unfocused, then ∼0.1% of the HY is photodissociated and the produced densities of the spin-polarized atoms are of the order of 10^{16} cm^{-3}. This is the low-density regime, where the depolarization mechanism is via the formation of a short-lived complex species between the spin-polarized hydrogen isotope and the hydrogen halide. The polarization lifetime is strongly dependent on the quadrupole moment of the halogen nucleus [10]. If the laser beam is focused, then almost all the HY molecules can be photodissociated near the focal point. As a result, the spin-polarized hydrogen isotope density approaches the initial HY density (10^{18}–10^{19} cm^{-3}) and the high-density regime is observed. In this case, the depolarization mechanism is dominated by collisions of polarized hydrogen isotopes with halogen atoms. The collision rate can be reduced by inserting an inert gas with a high heat capacity in order to cool down the spin-polarized hydrogen isotopes, and thus reduce the collision and depolarization rate.

4. Production and detection of polarized proton beams at the Forschungszentrum Jülich

The photodissociation method is adopted by M. Büscher's group for the production of polarized proton beams at the Forschungszentrum Jülich (FZJ) [12]. The laser system produces two spatially separated pulsed beams with wavelengths 1064 nm (fundamental) and 213 nm (5$^{\text{th}}$ harmonic). The 1064 nm beam with a pulse energy of ∼100 mJ is linearly polarized and focused into the interaction chamber in order to align the molecular bonds. The UV light pulses, which are circularly polarized, have an energy of ∼20 mJ and are used to photodissociate the molecules and produce the spin-polarized hydrogen atoms. The polarization oscillates between the electrons and the protons, so that half of the polarization is transferred to the nuclei after a long time ($t \gg 0.7$ ns), i.e. much longer than the inverse of the hyperfine frequency of H. Then, the spin-polarized H atoms get ionized and accelerated forming a proton beam, whose polarization can be measured by means of a Lamb-shift polarimeter [13]. The proton polarization is conserved during the beam passage through the detection scheme (ionizer, Wien filter, Cs cell, spin filter, and quenching chamber).

On the whole, molecular photodissociation is a method that can produce almost instantaneously and in situ high-density spin-polarized hydrogen isotopes. These features make it ideal for applications, such as laser acceleration of polarized particles and polarized laser fusion. Various experimental concepts have been proposed for either spin-polarized electron beams or proton/deuteron beams. Further applications, beyond the capabilities of conventional methods, are given elsewhere [8].

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