Polarizing nuclear-spin by a sequence of short-laser pulses: application to polarize muonium

Takashi Nakajima\textsuperscript{1}, Yukari Matsuo\textsuperscript{2} and Tohru Kobayashi\textsuperscript{2}

\textsuperscript{1} Institute of Advanced Energy, Kyoto University, Gokasho, Uji, Kyoto 611-0011, Japan
\textsuperscript{2} The Institute of Physical and Chemical Research (RIKEN) 2-1 Wako, Hirosawa, Saitama 351-0198, Japan
E-mail: nakajima@iae.kyoto-u.ac.jp

Abstract. We propose a new and simple scheme to polarize nuclear-spin by a sequence of short-laser pulses. This is a variant of optical pumping, but completely free from a complicated optimization procedure which is usually required to match the laser spectrum to that of the hyperfine transition lines of the target. Moreover the time needed to complete the polarization is in the order of tens of ns, which is more than a few orders of magnitude shorter than that usually needed to polarize nuclei with a continuous-wave (CW) laser. As a specific example we apply the idea to polarize muonium ($\mu^+e^-$, lifetime 2.2 $\mu$s).

1. Introduction

Developing an efficient technique to polarize nuclei is one of the important subjects in both nuclear physics and high energy physics. For that purpose there are a few well-known methods such as the nuclear-fragmentation method, optical pumping by a continuous-wave (CW) laser \cite{1, 2, 3, 4, 5, 6}, and a combination of optical pumping and spin-exchange collisions \cite{7, 8}. If a level structure of the target is simple and a powerful light source is available for a CW laser, optical pumping may be a good choice to polarize nuclei. However, if the pumping wavelength has to be as short as the vacuum-ultraviolet (VUV) wavelength, direct application of optical pumping with a CW light laser becomes practically impossible, since a bright light source is not available at such a short wavelength.

This is indeed the case of the muonium ($\mu^+e^-$) and hydrogen atom, etc. for which the pumping wavelength is in the VUV range, 122 nm (Lyman-$\alpha$ line). In particular polarizing the muonium is far more difficult than polarizing the hydrogen atoms, since the former has a very short lifetime (2.2 $\mu$s).

The purpose of this paper is to describe a new and simple scheme \cite{9} to polarize a target which cannot be polarized otherwise, and theoretically demonstrate its capability. We specifically choose the muonium as a target because there are no known methods to efficiently polarize the muonium regardless of the fact that the polarized muon is of great interest \cite{10, 11, 12} for many years. The key idea is to use a sequence of short laser pulses instead of a CW laser. This is a variant of optical pumping with an important difference that a short-pulse laser is used instead of a CW laser. Note that there are two important advantages of using a short-pulse laser over a CW laser for optical pumping, in particular in the VUV range. First, the efficiency of nonlinear frequency conversion, which is necessary to produce the VUV light, is much better with an
intense short laser pulse compared with a CW laser. Second, the spectral bandwidth of short laser pulses is extremely broad compared with those of CW and ns lasers, as a result of which we do not have to make efforts to match the laser spectral profile to the hyperfine transition lines of the target atom for the efficient pumping.

2. Model

In Fig. 1(a) we show the relevant level structure of the muonium atom. The level structure of the muonium ($\mu^+e^-$) is very similar to that of the hydrogen atom. The main difference between them can be seen in their hyperfine splittings. This is because the magnetic moment of the muon is much larger compared with the proton. Our assumption is that a sequence of laser pulses at a central photon energy of $\sim$10.2 eV with right-circular polarization coherently excites atoms in the ground $1s_{1/2}$ state to the excited $2p_{1/2}$ and $2p_{3/2}$ states which have a lifetime of 1.62 ns. Due to the presence of hyperfine interactions, each state splits into the two hyperfine levels, and Fig. 1(b) shows the level scheme of interest with all magnetic sublevels explicitly represented.

In this paper we assume that the duration of each pulse is 1 ps which has a spectral bandwidth of 440 GHz and the time interval between the pulses is several ns. Clearly if the time interval between the pulses is several ns, most atoms in the excited states go back to the ground state through the spontaneous decay before the next pulse arrives. Note that the dynamics of this system cannot be correctly described by rate equations due to the time-varying change of nuclear-spin orientation between the pulses. Moreover, a special care has to be taken for the present case where the hyperfine coupling time is comparable to the lifetime of the $2p$ state. Namely the use of the coupled basis represented by $\langle ((l s) j I) F M_F \rangle$ is not very convenient to correctly describe the entire process. Instead we use the uncoupled basis description, $\langle ((l s) j M_j) I M_I \rangle$, for the excited states [9]. After these considerations we employ a set of density matrix equations and numerically solve them to correctly describe the nuclear-spin dynamics.

3. Results and Discussions

Now we present some results. To start with we show the result for a single pump pulse. Figure 2 shows the variation of nuclear-spin as a function of laser detuning with different peak intensities, where zero detuning means that the 10.2 eV pump pulse is on resonant with the $1s_{1/2}$ ($F = 1$)-$2p_{1/2}$ ($F = 0$) hyperfine transition. Positive (negative) detunings imply that the photon energy is larger (smaller) than this. As the peak intensity increases the degree of spin-polarization increases up to $2 \times 10^8$ W/cm$^2$. If we further increase the peak intensity, for instance $4 \times 10^8$ W/cm$^2$,
the degree of spin-polarization starts to decrease because the *effective* pulse area (modulus of $\pi$) becomes smaller. Clearly we have no reason to make the peak intensity of each pulse higher than $2 \times 10^8$ W/cm$^2$.

Now we choose the detuning to be zero and increase the number of pulses to two. In Fig. 3 we show the change of spin-polarization for the different peak intensities as a function of time interval between the pulses. We observe the oscillations. This is essentially Ramsey interference in the time domain. Note that there are more than a few frequency components in the oscillations, since we are dealing with the multiple hyperfine transitions by short laser pulses with a very broad spectral bandwidth. Clearly the fastest oscillations are associated with the fine structure of $2p_{1/2}$ and $2p_{3/2}$, and hence $\sim 10.9$ GHz. Because of the presence of hyperfine levels in $1s_{1/2}$, $2p_{1/2}$, and $2p_{3/2}$, there are a few slightly different frequency components which result in the slow modulation in Fig. 3. Clearly if the time interval is made longer the influence of Ramsey interference becomes smaller due to the rapid loss of coherence through spontaneous decay, and at the 10 ns time interval the modulations are almost gone. Figure 3 demonstrates that the use of the two pump pulses improves the maximum degree of spin-polarization from $\sim 33\%$ for the single pulse to $\sim 53\%$ for the two pulses at the peak intensity of $2 \times 10^8$ W/cm$^2$.

For the technical reason a shorter time interval is more convenient. But if it is too short we
must worry about the interference effect. This means that we must make a compromise. Based on the result in Fig. 3, we choose the time interval to be 5 ns, and increase the number of pulses to eight. The detuning is set to zero again. The results are shown in Fig. 4. When the peak intensity is low, $10^7$ W/cm$^2$, we see the almost linear increase of the degree of spin-polarization after each pulse. This is very similar to the case of the ordinary optical pumping by a CW laser. When the peak intensity is higher, we can realize surprisingly high spin-polarization. For example, the spin-polarization is as high as 90% after interacting with eight pulses if the peak intensity is $10^8$ W/cm$^2$.

Upon production of muons they are known to be perfectly polarized. However, when they pick up electrons to form muonium atoms, the degree of polarization is reduced to about 50%. It is interesting to consider how much we can increase the degree of spin-polarization with the present scheme. Results for the eight pulses with a 5 ns time interval are presented in Fig. 5. In this case we can achieve 94% spin-polarization if we employ eight pulses.

4. Conclusions
In conclusion, we have proposed a new scheme to efficiently polarize nuclei, and presented specific examples for the muonium. Our scheme is a variant of optical pumping and it utilizes
a sequence of short laser pulses instead of a CW laser. We have found that the achievable degree of spin-polarization with the proposed scheme is quite high. The use of intense short laser pulses has a big advantage over the CW laser since the nonlinear frequency conversion to the UV-VUV wavelength range, which is needed to pump the muonium, can be much more efficient. Moreover, due to the very broad spectral bandwidth of short laser pulses all hyperfine as well as fine structure transition lines are simultaneously excited. This means that, unlike the optical pumping by a CW laser, no complicated optimization is necessary for our scheme. Our theoretical calculations for the unpolarized muonium atoms have shown that the degrees of spin-polarization can be as much as 33, 50, and 80 % by using a single, two, and five 10.2 eV pulses. If we can use eight pulses, it can be as much as 90 %.

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