Spin polarization of Doppler-broadened atoms by the broadband nanosecond and transform-limited picosecond laser pulses: case study for the muonium

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We study the spin polarization of Doppler-broadened atoms by broadband nanosecond (ns) pulses, and compare the polarization efficiency with that by the transform-limited picosecond (ps) pulses that have the same spectral bandwidth. Specific calculations are performed for the case of muonium with a set of density matrix equations and with rate equations using the uncoupled basis states. We find that the polarization efficiency with the broadband ns laser pulses is rather good, although it is not as good as that with the transform-limited ps pulses. Our results imply that, depending on the available laser system, we can use either broadband ns or transform-limited ps laser pulses to polarize almost any Doppler-broadened atoms. © 2012 Optical Society of America

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1. INTRODUCTION

Spin-polarized species are very useful tools for investigating various kinds of interaction dynamics, not only in atomic physics, but also in nuclear physics [1] and high energy physics [2]. In particular, there is a strong demand to find an efficient scheme to polarize the nuclear spin of *unstable* species with a lifetime of milliseconds or even microseconds, which are of great interest in recent years and far more difficult to polarize than stable ones [1]. Among them, the muonium (μ^+e^-) is one of the most difficult species to polarize due to its short lifetime (2.2 μs) before annihilation. Our specific choice of muonium in this paper stems from the interest of the muon community [3] to produce highly spin-polarized ultraslow [sub-electron-volt (sub-eV)] muoniums for various applications. Ultraslow muoniums are obtained from high-energy [a few mega-electron-volts (MeV)] muons through the following processes.

Upon birth, muons (μ^+) are 100% spin polarized. For various applications, such as muon spin rotation (μSR) [4,5], the kinetic energy of muons must be greatly reduced down to the sub-eV energy [6,7]. For this purpose, a thin tungsten film is generally used [6]. When muons pass through the tungsten film, they capture electrons and become muoniums with a sub-eV energy at the expense of the loss of spin polarization down to about 50% [7]. Since the quality of the spin-polarized beam is usually defined by the figure of merit, P^2I, where P is a degree of spin polarization and I the flux or current of the beam, how to efficiently excite and finally ionize muoniums is a very important issue [6,7]. If the restoration of spin polarization is also achieved, it is even better.

The use of lasers is a strong candidate to resonantly excite, polarize, and finally ionize muoniums. However, things are not so simple. First, the transition wavelength we need to resonantly excite muoniums is 122 nm, which is in the vacuum-ultraviolet (Lyman-α line). Although there are many attempts to produce Lyman-α light [8–12], there is no well-established scheme to obtain nanosecond (ns) Lyman-α pulses with sufficient energy. The brightness of Lyman-α in the continuous wave (CW) mode [13,14] would not be enough, either. Second, the ultraslow muoniums, after passing through the tungsten film, are Doppler-broadened as much as ~230 GHz due to the temperature of the tungsten film. This is a huge amount of Doppler width, not only for CW lasers but also for ns lasers. Recall that the spectral width of the transform-limited (i.e., perfectly coherent) laser pulse is given by the inverse of the pulse duration due to the uncertainty principle, and it is a few hundred megahertz (MHz) [gigahertz (GHz)] for a few ns [picosecond (ps)] laser pulses.

We have recently shown that the use of a sequence of ps Lyman-α pulses can solve all these problems [15]. Because the spectral width of ps pulses is comparable to the Doppler width of ultraslow muoniums, we can efficiently excite/polarize ultraslow muoniums by the sequence of ps Lyman-α pulses. In response to our proposal [15], there is a question addressed by the muon community that the use of ns Lyman-α pulses with the same (~230 GHz) bandwidth may do the similar job. If this is indeed the case, it is a very good news for experimentalists, since one can use either broadband ns or transform-limited ps pulses, depending on the available laser system. We should not forget, however, that the broadband ns pulses have poorer coherence than the transform-limited ps pulses with the same bandwidth, and this difference would affect the polarization efficiency. Therefore, how much spin polarization we can achieve by using the broadband ns pulses is not clear without quantitative investigation. Note that this kind of comparison for the polarization efficiency by the ns and ps laser pulses with the same spectral bandwidth does not exist in the literature.

In this paper, we study spin polarization of Doppler-broadened muoniums by the broadband ns Lyman-α pulses, and compare the polarization efficiency with that by transform-limited ps Lyman-α pulses that have the same spectral bandwidth. We employ a set of density matrix equations and also rate equations to calculate spin polarization by the broadband ns Lyman-α pulses. As we will show in this paper, we find that the broadband ns pulses can rather efficiently polarize muoniums, although the efficiency is not as good as that by the transform-limited ps pulses with the same bandwidth. The data we report in this paper can serve as a useful benchmark to develop a suitable Lyman-α light source to polarize Doppler-broadened muoniums. Perhaps even more importantly, our findings are obviously quite general and, hence, very useful not only to polarize muoniums but also almost any kind of atom [16–18]. That is, our results suggest that, as long as the spectral bandwidth of the pump laser covers the Doppler broadening of target atoms, the degree of coherence of the pump source is of secondary importance, and we can obtain high polarization efficiency for most atoms by a sequence of broadband (and, hence, only partially coherent) ns laser pulses. We would like to emphasize that the broadband ns laser pulses are much easier to handle than the high-power CW laser, which requires precise tuning of the pumping wavelength or transform-limited laser pulses, which, in turn, requires a sophisticated arrangement for the lasing process.

2. MODEL

The level scheme we consider in this paper is identical with that described in Fig. 1 of our previous paper [15], where the *uncoupled basis states* have been employed to describe the spin-polarization dynamics induced by a sequence of ps laser pulses. Note that we had to use the uncoupled basis states in the recent paper [15] due to the slow hyperfine coupling time (several ns) of the muonium $2p$ hyperfine states, and the same is true for the broadband ns laser pulses. Since the ns laser pulses we assume in this paper have a broad bandwidth, one may naively think that the use of ordinary rate equations may be valid. This is not correct. Recall that the existing rate equation analysis work for optical pumping by the CW laser utilizes the ordinary coupled basis states [19], since the CW laser can resolve the hyperfine structure. We point out that a similar rate equation analysis for the ns laser pulses with the uncoupled basis states has never been undertaken to describe the temporal change of spin polarization. The relevant states described in Fig. 1(b) are $|0\rangle = |(1s_{1/2})F=0M_F=0\rangle$, $|6\rangle = |(1s_{1/2})F=1M_F=0\rangle$, $|7\rangle = |(2p_{1/2})M_J=1/2\rangle|M_I=1/2\rangle$, $|8\rangle = |(2p_{3/2})M_J=1/2\rangle|M_I=1/2\rangle$, $|9\rangle = |(2p_{3/2})M_J=3/2\rangle|M_I=-1/2\rangle$, $|10\rangle = |(1s_{1/2})F=1M_F=1\rangle$, and $|11\rangle = |(2p_{3/2})M_J=3/2\rangle|M_I=1/2\rangle$. If we define $\sigma_{jj}(j=0,6-11)$ as the population of state $|j\rangle$, the rate equations of the system in the uncoupled basis states read

$$\dot{\sigma}_{00} = \gamma_{sp}\left(\frac{1}{3}\sigma_{77} + \frac{1}{6}\sigma_{88} + \frac{1}{2}\sigma_{99}\right) - \sum_{k=7,8,9}\frac{\gamma\Omega_{0k}^2}{\Delta_{k0}^2 + (\gamma/2)^2}(\sigma_{00} - \sigma_{kk}), \quad (1)$$

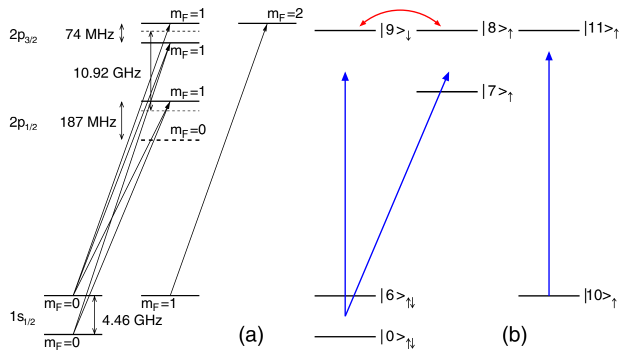

Fig. 1. (Color online) (a) Level scheme considered in this paper where the muonium is initially 50% spin polarized. (b) Relevant states in terms of the uncoupled basis description. The curved arrow between $|8\rangle$ and $|9\rangle$ indicates hyperfine coupling, and the up/down arrows by the ket vectors represent the nuclear-spin orientations. Note that the numbering of the states is not sequential, to be consistent with the numbering of states in our previous work [15], where a set of density matrix equations have been derived.

$$\dot{\sigma}_{66} = \gamma_{sp}\left(\frac{1}{3}\sigma_{77} + \frac{1}{6}\sigma_{88} + \frac{1}{2}\sigma_{99}\right) - \sum_{k=7,8,9}\frac{\gamma\Omega_{6k}^2}{\Delta_{k6}^2 + (\gamma/2)^2}(\sigma_{66} - \sigma_{kk}), \quad (2)$$

$$\dot{\sigma}_{77} = -\gamma_{sp}\sigma_{77} + \sum_{k=0,6}\frac{\gamma\Omega_{k7}^2}{\Delta_{7k}^2 + (\gamma/2)^2}(\sigma_{kk} - \sigma_{77}), \quad (3)$$

$$\dot{\sigma}_{88} = -\gamma_{sp}\sigma_{88} + \sum_{k=0,6}\frac{\gamma\Omega_{k8}^2}{\Delta_{8k}^2 + (\gamma/2)^2}(\sigma_{kk} - \sigma_{88}) + \frac{2\Omega_{89}^{(H)2}}{\gamma_{sp}}(\sigma_{99} - \sigma_{88}), \quad (4)$$

$$\dot{\sigma}_{99} = -\gamma_{sp}\sigma_{99} + \sum_{k=0,6}\frac{\gamma\Omega_{k9}^2}{\Delta_{9k}^2 + (\gamma/2)^2}(\sigma_{kk} - \sigma_{99}) - \frac{2\Omega_{89}^{(H)2}}{\gamma_{sp}}(\sigma_{99} - \sigma_{88}), \quad (5)$$

$$\dot{\sigma}_{1010} = \gamma_{sp}\left(\frac{1}{3}\sigma_{77} + \frac{2}{3}\sigma_{88} + \sigma_{1111}\right) - \frac{\gamma\Omega_{1110}^2}{\Delta_{1110}^2 + (\gamma/2)^2}(\sigma_{1010} - \sigma_{1111}), \quad (6)$$

$$\dot{\sigma}_{1111} = -\gamma_{sp}\sigma_{1111} + \frac{\gamma\Omega_{1110}^2}{\Delta_{1110}^2 + (\gamma/2)^2}(\sigma_{1010} - \sigma_{1111}), \quad (7)$$

where Ω_{ij} is a Rabi frequency between states $|i\rangle$ and $|j\rangle$, $\Omega_{89}^{(H)}$ is the hyperfine coupling between states $|8\rangle$ and $|9\rangle$, and Δ_{ij} is a laser detuning defined by $\Delta_{ij} \equiv \omega - (\omega_i - \omega_j)$, in which ω_i is the energy of state $|i\rangle$. γ_{sp} and γ_L are the spontaneous decay rate, which is 1.6 ns for the $2p$ state and laser bandwidth, respectively, with $\gamma \equiv \gamma_{sp} + \gamma_L$. Note that γ_L represents the bandwidth arising from the incoherent processes, such as phase fluctuations of the laser field [20], and can be written as $\gamma_L = \gamma_{L0} b^2/(\Delta_{ij}^2 + b^2)$, in which the cutoff parameter, b, has been introduced to correct the Lorentzian laser bandwidth, γ_{L0}; otherwise, we will obtain unphysical results for the off-resonant case due to the very slow fall-off of the Lorentzian laser spectrum. In this paper, we choose $\gamma_{L0} = 230$ GHz with $b = 0.8\gamma_{L0}$ and $\gamma_{L0} = 0$ GHz for the broadband 2 ns and transform-limited 2 ps laser pulses, respectively, with a Gaussian temporal shape. As for the initial condition, we assume $\sigma_{00} = \sigma_{66} = 1/4$, and $\sigma_{1010} = 1/2$ at $t = -\infty$, which is actually the case when muoniums are born after muons pass through the tungsten film. The ionization cross section from the $2p$ state by Lyman-α light is about 0.5 Mb, and, hence, we estimate that the ionization probability from the $2p$ state is negligibly small ($<10^{-4}$) for the pulse durations and intensities we consider in this paper.

After solving the above equations, we can calculate the degree of spin polarization, P, from the relation of

$$P = \frac{P_{\mathrm{up}} - P_{\mathrm{down}}}{P_{\mathrm{up}} + P_{\mathrm{down}}}, \tag{8}$$

where

$$P_{\mathrm{up}} = \frac{1}{2}\sigma_{00}(t=\infty) + \frac{1}{2}\sigma_{66}(t=\infty) + \sigma_{1010}(t=\infty), \tag{9}$$

$$P_{\mathrm{down}} = \frac{1}{2}\sigma_{00}(t=\infty) + \frac{1}{2}\sigma_{66}(t=\infty). \tag{10}$$

3. RESULTS AND DISCUSSION

In Fig. 2 we show the total population of the excited states in the absence of Doppler broadening as a function of laser detuning by the broadband 2 ns and transform-limited 2 ps laser pulses after appropriate normalization of the vertical scale. Clearly, the two laser pulses have very similar spectral shapes with 230 GHz bandwidths [full width at half-maximum (FWHM)], with a small difference in the wings. This difference is inevitable, since the physical origin of the laser bandwidths for the ns and ps pulses is completely different. However, this slight difference in the laser spectral shapes does not spoil the following discussions and conclusions we will draw, since most of the pumping dynamics take place within the laser bandwidths.

Now we present the results of spin polarization by the broadband 2 ns pulses. Using Eqs. (1)–(34) in the recent paper [15] together with the additional laser bandwidth terms, $-(\gamma/2)\sigma_{ji}$, to be added to the equations for the off-diagonal terms, σ_{ji}, connecting the states $|i\rangle$ and $|j\rangle$ ($i = 0$, 6, 10; $j = 7$, 8, 9, and 11), we carry out the calculations for muoniums under the Doppler broadening of 230 GHz (FWHM) with a Gaussian distribution function. To test the validity of rate equation description, we have also shown the results by solving the rate equations given by Eqs. (1)–(7). The results are presented in Fig. 3. Figure 3(a) shows the variation of spin

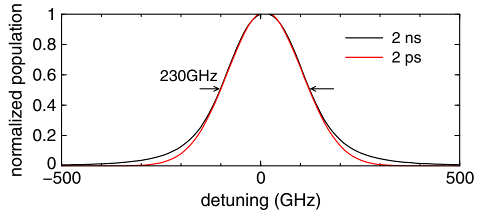

Fig. 2. (Color online) Excited state population as a function of laser detuning by the 2 ns pulse with a 230 GHz bandwidth and transform-limited 2 ps laser pulse. Doppler broadening is not included in the calculations.

polarization after interacting with a single 2 ns pulse at the intensities of 10^4, 10^5, and 10^6 W/cm^2. Clearly, if the intensity is sufficiently high, we can restore spin polarization from 50% to over 70% by the irradiation of the single 2 ns pulse, even under the presence of 230 GHz Doppler width. If we use a sequence of 2 ns pulses, it is even better, as we show in Fig. 3(b). For example, if we use four 2 ns pulses at the intensity of 10^5 W/cm^2 with a time interval of 5 ns, spin polarization increases from 50% to over 80%.

Similar results are shown in Figs. 4(a) and 4(b) for the transform-limited 2 ps pulses. The reduction of spin polarization in Fig. 4(b) after the intensity of 4×10^7 W/cm^2 is due to the Rabi oscillation: Once the complete population inversion takes place at the intensity of about 4×10^7 W/cm^2, the pump efficiency decreases until it reaches the null pumping efficiency, and so on. Note that the Rabi oscillation does not take place for the broadband 2 ns pulses, and, hence, spin polarization monotonically improves as the intensity increases [Fig. 3(b)]. Indeed this is the reason why the dynamics by the broadband 2 ns pulses are well described by the rate equations.

It is interesting to note that, given the same total pulse energy, the use of a sequence of pulses can attain higher spin polarization than the single pulse with higher intensity. For instance, see Fig. 3(b) around 10^4 W/cm^2. By using the single 2 ns pulse at the peak intensities of 2×10^4 and 4×10^4 W/cm^2,

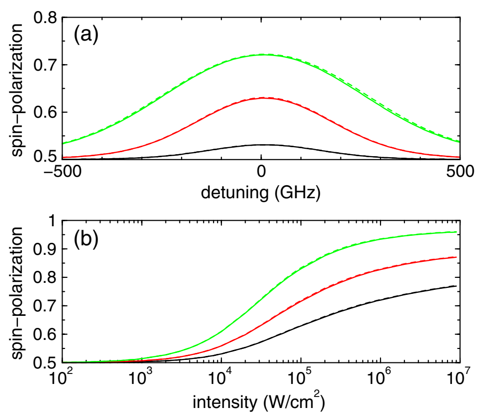

Fig. 3. (Color online) (a) Spin polarization as a function of laser detuning by the single 2 ns laser pulse at the intensities of 10^4 (lower curve), 10^5 (middle curve), and 10^6 W/cm^2 (upper curve). (b) Spin polarization as a function of intensity by the one (lower curve), two (middle curve), and four (upper curve) 2 ns laser pulses. The laser bandwidth is 230 GHz for all curves. In both panels (a) and (b), the thick and thin curves represent the results with and without 230 GHz Doppler broadening, and the dashed curves are the results obtained by solving the rate equations.

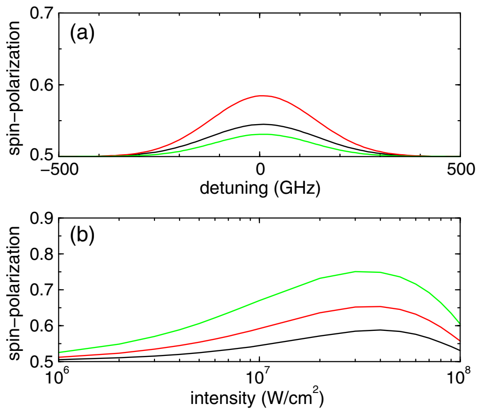

Fig. 4. (Color online) (a) Spin polarization as a function of laser detuning by the single 2 ps laser pulse at the intensities of 10^7 (middle curve), 5×10^7 (upper curve), and 10^8 W/cm^2 (lower curve). (b) Spin polarization as a function of intensity by the one (lower curve), two (middle curve), and four (upper curve) 2 ps laser pulses. The 2 ps pulses are transform limited for all curves. In panels (a) and (b), the thick and thin curves represent the results with and without 230 GHz Doppler broadening.

we can obtain spin polarization of 55% and 58%. In contrast, if we use two and four 2 ns pulses at the intensity of 10^4 W/cm^2, spin polarization is even higher, 56% and 61%. The similar is true for the case of 2 ps pulses. These findings suggest that a careful consideration, which includes the energy loss to split the pulses, would be necessary in the real experiment to estimate in advance whether the single or a sequence of pulses can be more efficient.

Now we compare the polarization efficiency by the use of the broadband 2 ns and transform-limited 2 ps pulses. To make a concrete argument, we assume the same beam diameter and pulse energy for the 2 ns and 2 ps Lyman-α pulses. This means that the comparison should be made after upshifting the intensity scale of the results by the 2 ns pulses by 3 orders of magnitude; that is, the results at 10^3 W/cm^2 for the 2 ns pulses should be compared with those at 10^6 W/cm^2 for the 2 ps pulses. The comparison is made in Fig. 5 by superposing Fig. 3(b) over Fig. 4(b). We can see that, up to the intensity of 4×10^7 W/cm^2, where spin polarization for the 2 ps pulse reaches the maximum, the use of 2 ps pulses is more efficient than the 2 ns pulses to polarize the muonium. This, however, does not mean that the 2 ps pulses are more efficient only in the certain intensity range. Actually, we can always control the intensity of the 2 ps pulses by splitting the pulses or increasing the beam diameter, etc., so that their intensities are kept below $\sim 3 \times 10^7$ W/cm^2, where the 2 ps pulses are always more efficient than the 2 ns pulses. Another thing we can find from Fig. 5 is that the superiority of the use of 2 ps pulses becomes more obvious as the number of pulses increases.

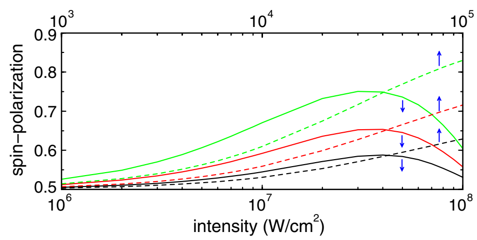

Fig. 5. (Color online) Comparison of spin-polarization efficiency by the 2 ns and 2 ps pulses. The results by the single, two, and four pulses with a duration of 2 ps (2 ns) with the time interval of 5 ns are shown by the solid (dashed) lower, middle, and upper curves, respectively.

4. CONCLUSIONS

In summary we have investigated spin polarization of Doppler-broadened muoniums by broadband 2 ns pulses, and compared the polarization efficiency with that by transform-limited 2 ps laser pulses that have the same spectral bandwidth, 230 GHz. Since the hyperfine coupling time of the excited states is comparable to the spontaneous decay time, the use of the coupled basis states is not adequate to describe the dynamics, and, therefore, we have employed uncoupled basis states for both density matrix and rate equations. The results obtained by the rate equations for the broadband ns laser pulses agree quite well with those by the set of density matrix equations. We have found that the efficiency to polarize Doppler-broadened muoniums by the broadband ns pulses is rather good, although it is not as good as that by the transform-limited ps pulses. Our results imply that, as long as the spectral bandwidth of the pump laser covers the Doppler broadening of target atoms, the degree of coherence of the pump source is of secondary importance, and we can use either broadband ns or transform-limited ps laser pulses to polarize almost any Doppler-broadened atoms, which greatly increases the applicability of the optical pumping technique for various purposes.

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