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Nuclear-spin polarimeter using photoelectron signals

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It is widely believed that the hyperfine states must be resolved in one way or the other to measure the degree of nuclear-spin polarization of atoms and ions. We theoretically show that the angle-resolved photoelectron signals produced by resonant multiphoton ionization by short and hence broadband two-color laser pulses, which cover all fine-structure and hyperfine manifold of the ground and resonant excited states, can serve as a new type of nuclear-spin polarimeter. Because of the use of time-delayed short laser pulses the proposed method has very high (subnanosecond) time resolution, and does not suffer from the Doppler broadening. Specific results are presented for the two-photon resonant three-photon ionization of hydrogen atoms using 205- and 410-nm laser pulses with picosecond to femtosecond pulse durations. The proposed scheme is immediately applicable to other unstable elements such as $^{11}\mathrm{Be}^+$, $^{27}\mathrm{Mg}^+$, and $^{35}\mathrm{Ca}^+$, etc.

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I. INTRODUCTION

The last few decades have witnessed the broad applications of spin-polarized nuclei which range from basic science such as high-energy physics and nuclear physics [1] to medical science utilizing NMR, and the rapidly growing area of spintronics [2].

In spite of its usefulness, neither the production nor the detection of nuclear-spin polarization is an easy task. So far, only a few methods have been employed to produce nuclear-spin polarized species. These methods include nuclear fragmentation, which is mostly used in nuclear physics experiments, photodissociation of molecules [3,4], Stern-Gerlach separation [5], optical pumping [6–8], and spin-exchange optical pumping [9]. When it comes to the methods of detecting the degree of nuclear-spin polarization, the known methods are even fewer. The β-NMR method is very useful, but it works only for unstable nuclei with β decay [10]. Besides, its temporal resolution is limited by the lifetime of the unstable nuclei. We note that there is no well-established method to detect nuclear-spin polarization of stable nuclei other than hydrogen atoms.

Even for hydrogen atoms the detection is technically complicated, and large detectors of proton polarimeters [11] and Lamb-shift polarimeters [12,13] have been employed for many years.

More recently, optical detection of (nuclear-)spin polarized hydrogen or other similar atoms or ions through laser-induced fluorescence (LIF) and resonantly enhanced multiphoton ionization (REMPI) [14–17] have been demonstrated. The LIF method works well at a relatively high atomic density, and accordingly, the detection of LIF down to the ground state is not very appropriate due to the possible occurrence of radiation trapping. Of course, the detection of LIF between the two excited states would not cause such a problem, but then, the time resolution would not be better than several nanoseconds due to the longer spontaneous lifetime. The above feature of the LIF method implies that it is perhaps more suitable to monitor spin polarization of the atomic gas target rather than that of the atomic beam. As for the REMPI method, it has a better detection sensitivity and works well at a lower atomic density. This implies that the REMPI method may be more suitable to monitor spin polarization of an atomic beam. However, the use of narrow-band lasers is required to spectrally resolve the fine structure of the resonant excited state, and consequently, this method easily suffers from the Doppler broadening. The requirement that the laser bandwidth be sufficiently narrow also implies that the time resolution is rather limited to several nanoseconds.

In this paper we theoretically show that the degree of nuclear-spin polarization can be realiably measured by the angle-resolved photoelectron signals [18] which are produced by resonantly enhanced two-step multiphoton ionization. As an example, we choose one of the most popular and commonly used atoms, the hydrogen atom, to demonstrate the performance of the proposed scheme. Because of the similarity of the electronic structure and the value of nuclear spin, $I = 1/2$, our scheme is also immediately applicable to some other atoms such as $^{11}\mathrm{Be}^+$, $^{27}\mathrm{Mg}^+$, and $^{35}\mathrm{Ca}^+$, which are unstable nuclei and are of recent interest in nuclear physics. We would like to stress that the *production* of nuclear-spin-polarized species by two-step photoionization [19] is not the same as the *detection* of nuclear-spin polarization by two-step photoionization that we will report in this paper. Actually, the latter is much more demanding, and indeed we do not find any work in the literature which discusses the possibility of using photoelectron signals as a nuclear-spin polarimeter.

Before we go into the detail, we make a few remarks on the difference between previously mentioned LIF and REMPI methods and our scheme. Our scheme is in some sense similar to the REMPI method and works well under the low atomic density, and hence is more suitable to analyze spin polarization of an atomic beam for various kinds of collision experiments. Its advantage over the REMPI method is that it does not suffer from the Doppler broadening and has a higher time resolution, subnanosecond. As we mentioned before, the working conditions for our scheme and LIF methods are different and they would be suitable for different purposes.

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To be more specific, we consider two-photon resonant, three-photon ionization of hydrogen atoms by time-delayed 205- and 410-nm pulses with short (approximately 10 ps to 100 fs) pulse durations. Note that the equations and results we will show later in this paper do not depend on the specific choice of pulse duration, as long as the corresponding laser bandwidth is much broader than the fine-structure splitting of the resonant excited state and Doppler width but much narrower than the energy intervals between the resonant and other electronic states.

II. TWO-PHOTON RESONANT THREE-PHOTON IONIZATION THROUGH THE 3*d* MANIFOLD

Before we arrived at the present scheme, we considered the single-photon ionization process of hydrogen atoms directly from the hyperfine ground states and found that the photoelectron angular distribution (PAD) is always identical, regardless of the degree of nuclear-spin polarization of atoms in the hyperfine ground states. We would like to emphasize that our scheme requires spectral resolution of neither hyperfine nor fine-structure states. Actually the requirement is the other way around: In order for the present scheme to work as a nuclear-spin polarimeter, it is essential that the fine-structure states as well as all the relevant hyperfine states are coherently excited before photoionization, which is somehow counterintuitive. In this sense, the first (pump) pulse may be a pulse to couple 1*s* and 2*p* states. The wavelength, however, is 121.6 nm in the vacuum ultraviolet region, and it is not technically convenient. A much more convenient choice is a pump pulse at 205 nm with right-circular polarization to couple 1*s* and 3*d* states by two-photon excitation. The second (probe) pulse can be at any wavelength, but we can most conveniently choose 410 nm with linear or circular polarization.

The scheme we consider is shown in Fig. 1. A short pump pulse at 205 nm with right-circular polarization excites atoms from the 1*s* manifold to the 3*d* manifold by a two-photon transition. A time-delayed short probe pulse at 410 nm induces photoionization from the 3*d* manifold. Of course the time delay must be smaller than the natural lifetime of the excited states, which is about 11.5 ns for the 3*d* state. In order to coherently excite all (fine-structure and hyperfine) states belonging to the 3*d* manifold (see Fig. 1), we assume that both pulses have pulse durations of 10 ps to 100 fs, whose bandwidths are sufficiently broader than the fine-structure splitting, which is ∼1 GHz for the 3*d* manifold, but sufficiently narrow so that the pump pulse does not excite other undesired electronic states. As we mentioned before, a precise choice of the pulse durations does not influence the equations and results we present in this paper, as long as the above conditions for the pulse durations or equivalently, the laser bandwidths are satisfied. It is worth noting that, because of the presence of hyperfine interaction, the 1*s* manifold can be coupled to the 3*s* manifold even if the pump pulse is right-circularly polarized; for instance, the transition probability of $1s\ {}^2S_{1/2}\ (F=1,m_F=-1) \rightarrow\rightarrow 3s\ {}^2S_{1/2}\ (F=1,m_F=1)$ by two-photon excitation is not exactly zero. However, it turns out that the coupling strength is extremely weak compared with the other transitions from 1*s* to 3*d* manifold, and hence we may neglect the entire 3*s* manifold. In the following analysis we assume that both pump and probe pulses are sufficiently weak so that we may employ the perturbation theory for the excitation and ionization processes.

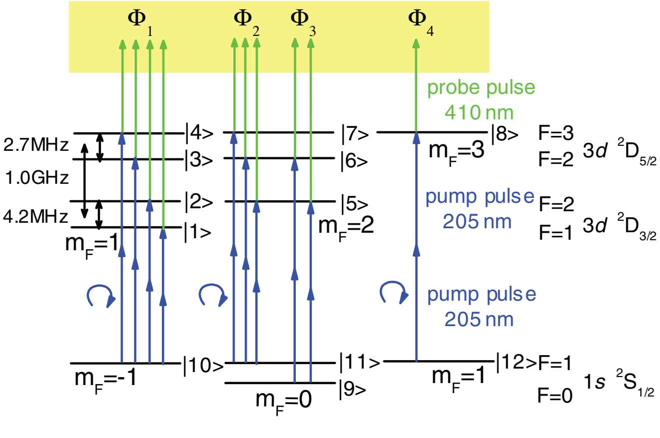

FIG. 1. (Color online) Level scheme of hydrogen atoms we consider in this paper. A short pump pulse at 205 nm with right-circular polarization excites atoms in the 1*s* manifold to the 3*d* manifold by two-photon absorption, and a time-delayed short probe pulse at 410 nm with right-circular, left-circular, or linear polarization induces photoionization.

For hydrogen atoms (nuclear spin of $I = \frac{1}{2}$), we define the degree of nuclear-spin polarization as

$$P_{\mathrm{H}} = \frac{P_\uparrow - P_\downarrow}{P_\uparrow + P_\downarrow}\ , \tag{1}$$

where $P_\uparrow$ and $P_\downarrow$ are the populations of atoms in the hyperfine ground states with nuclear-spin up and down, respectively. They read

$$P_\uparrow = |C_{12}|^2 + \tfrac{1}{2}(|C_9|^2 + |C_{11}|^2) \tag{2}$$

and

$$P_\downarrow = |C_{10}|^2 + \tfrac{1}{2}(|C_9|^2 + |C_{11}|^2), \tag{3}$$

in which C_j $(j = 9,10,11,12)$ is a probability amplitude of atoms in state $|j\rangle$ (see Fig. 1). The above definition of P_{H} implies that the quantity we must measure is actually the population difference of ground-state atoms with *electron-spin* up and down, which is practically identical to that with *nuclear-spin* up and down. This is the reason why the spin-orbit interaction rather than the hyperfine interaction of the excited state plays a major role for our scheme. Obviously, depending on the initial state, $|j\rangle$ $(j = 9,10,11,12)$, a different superposition of excited states is produced by the pump pulse and we call it Φ_k $(k = 1,2,3,4)$, which consists of relevant fine-structure and hyperfine states belonging to the 3*d* configuration. They are written as

$$\Phi_1 = C_{10}\left(\frac{\sqrt{2}}{5}|1\rangle + \frac{\sqrt{6}}{15}|2\rangle e^{-i\Delta\omega_{12}t} + \frac{2}{15}|3\rangle e^{-i\Delta\omega_{13}t} + \frac{\sqrt{2}}{15}|4\rangle e^{-i\Delta\omega_{14}t}\right),$$

$$\Phi_2 = C_{11}\left(\frac{2}{5\sqrt{3}}|5\rangle + \frac{2\sqrt{2}}{15}|6\rangle e^{-i\Delta\omega_{56}t} + \frac{\sqrt{2}}{3\sqrt{5}}|7\rangle e^{-i\Delta\omega_{57}t}\right),$$

$$\Phi_3 = C_9\left(-\frac{2}{5\sqrt{3}}|5\rangle + \frac{\sqrt{2}}{5}|6\rangle e^{-i\Delta\omega_{56}t}\right),$$

$$\Phi_4 = \frac{\sqrt{2}}{\sqrt{15}}C_{12}|8\rangle, \tag{4}$$

where $\Delta\omega_{ij}$ is the energy difference between states $|i\rangle$ and $|j\rangle$ and t is a time delay between the pump and probe pulses. By introducing the continuum state as $|\Psi\rangle$, we can write the yield of photoelectrons, I_e, flying away to the direction of (θ,φ) with respect to the quantization axis, which is chosen along the propagation direction for the circularly polarized pump pulse. It reads [19]

$$I_e(\theta,\varphi,t) \propto \sum_{k=1}^{4}\sum_{m_I,m_s} |\langle\Psi(\theta,\varphi)|D_q|\Phi_k(t)\rangle|^2, \tag{5}$$

where D_q is the qth spherical component of the dipole operator in which q takes the value of 0 or ±1 for the linearly or right- or left-circularly polarized probe pulse, respectively. For convenience we do the partial wave expansion of the continuum state, $|\Psi(\theta,\varphi)\rangle$, corresponding to the photoelectrons having the wave vector $\vec{k}$ and electron-spin orientation m_s with a nuclear-spin orientation m_I of the core (proton). Thus $|\Psi(\theta,\varphi)\rangle$ is expanded as

$$|\Psi(\theta,\varphi)\rangle = \sum_{l,m_l} a_{lm_l}|\vec{k};\ l\ m_l\rangle|s\ m_s\rangle|I\ m_I\rangle, \tag{6}$$

where $a_{lm_l} = 4\pi i^l e^{-i\delta_l} Y^*_{lm_l}(\theta,\varphi)$, with $Y_{lm_l}(\theta,\varphi)$ being the spherical harmonics. For hydrogen atoms, the phase shift δ_l of the l partial wave originates from the Coulomb phase shift only, and $\delta_l = \arg[\Gamma(l+1-i/\sqrt{W})] - \pi l/2$, with W being the kinetic energy of the photoelectrons in Rydberg units.

Using Eqs. (1)–(6) we calculate the PADs by the right-circularly polarized pump and linearly or right- or left-circularly polarized probe pulses for different values of P_H's. Although we can carry out numerical calculations very easily, it is useful to introduce some approximations to obtain the results in a closed form. By recalling that the lifetime of the $3d$ state is 11.5 ns and the spin-orbit and hyperfine coupling times are about 1 ns and a few hundred nanoseconds, respectively, it is safe to assume that the hyperfine coupling does not take place before atoms in the $3d$ state spontaneously decay. That is, $\cos\Delta\omega_{12}t = \cos\Delta\omega_{34}t = \cos\Delta\omega_{67}t \approx 1$ and $\cos\Delta\omega_{13}t = \cos\Delta\omega_{14}t = \cos\Delta\omega_{23}t = \cos\Delta\omega_{24}t = \cos\Delta\omega_{56}t = \cos\Delta\omega_{57}t \approx \cos\Delta\omega t$, where $\Delta\omega$ is the fine-structure splitting between $3d\ ^2D_{3/2}$ and $^2D_{5/2}$.

A. Right-circularly polarized probe pulse

With the above approximations for the various cosine terms, we obtain the expression for the PAD after very long and tedious algebraic calculations. It reads

$$\begin{aligned} I_e(\theta,t) \propto\ & \frac{8\pi}{25}f_1(\theta)(1-\cos\Delta\omega t)(1-P_H) \\ & + \frac{\pi}{100}f_2(\theta)(17+8\cos\Delta\omega t)(1-P_H) \\ & + \frac{\pi}{4}f_2(\theta)(1+P_H), \end{aligned} \tag{7}$$

where $f_1(\theta)$ and $f_2(\theta)$ are defined as

$$f_1(\theta) = \sin^4\theta\cos^2\theta\left|R^{kf}_{3d}\right|^2 \tag{8}$$

and

$$f_2(\theta) = \sin^6\theta\left|R^{kf}_{3d}\right|^2, \tag{9}$$

in which R^{kf}_{3d} is a radial matrix element for the bound-free transition from the $3d$ state to the kf continuum state. φ has been dropped from the arguments of I_e, since I_e has no dependence on φ under the current conditions. It takes the value of $R^{kf}_{3d} = 2.83$ (atomic units) for the probe pulse at 410 nm. Since we are assuming a weak probe pulse, the loss of population from the $3d$ manifold by photoionization is negligible. Besides, the loss of population and coherence from the $3d$ manifold by the spontaneous decay can also be neglected, since the spontaneous decay rate is the same for all the $3d$ manifold, and the time delay we consider in this paper is less than 1 ns, which is much smaller than the natural lifetime of the $3d$ state (11.5 ns). In order to ensure that there is no mistake in the calculations, we have carried out calculations using both a coupled and uncoupled basis description of the states [8], and obtained identical expressions.

Using Eq. (7), we plot the variation of the photoelectron signal as functions of θ and P_H. Results are shown in Figs. 2(a) and 2(b) at two different time delays between the pump and probe pulses. If $t = 0$ [Fig. 2(a)], the PAD shows no dependence on the value of P_H since the spin-orbit interaction, not to mention the hyperfine interaction, is not yet acting on the system. This situation is equivalent to the direct three-photon ionization by short laser pulses, with or without resonant intermediate states. In contrast, if $t = \pi/\Delta\omega$ [Fig. 2(b)], the PAD exhibits a significant dependence on P_H, since the largest amount of momentum transfer takes place at this moment

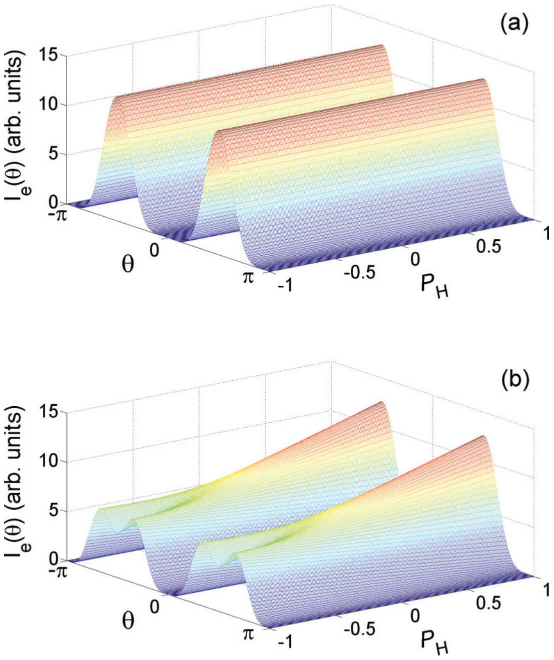

FIG. 2. (Color online) Variation of photoelectron signals $I_e(\theta,t)$ as functions of ejection angle θ and the degree of nuclear-spin polarization P_H at (a) $t = 0$ and (b) $t = \pi/\Delta\omega$.

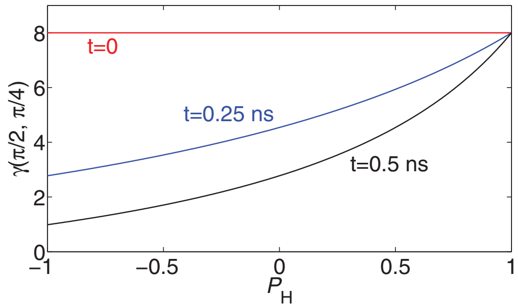

FIG. 3. (Color online) Relation between $\gamma(\frac{\pi}{2},\frac{\pi}{4})$ and $P_{\rm H}$ at the time delays of 0 ns (red), 0.25 ns (blue), and 0.5 ns (black), which correspond to $t=0$, $0.5\pi/\Delta\omega$, and $\pi/\Delta\omega$, respectively. The solid and dashed lines are the results with and without approximation to represent $\Delta\omega_{ij}$.

from the orbital component to the electron-spin component of the $3d$ state through spin-orbit interaction, where the nuclear-spin component is kept frozen. This implies that the PAD can be used as a new type of nuclear-spin polarimeter to determine $P_{\rm H}$.

Experimentally the easiest way to measure $P_{\rm H}$ is to take the ratio of photoelectron signals measured at two different θ values and correlate it to $P_{\rm H}$. We choose $\theta=\pi/4$ and $\pi/2$, and take the ratio of the corresponding photoelectron signals to obtain

$$\gamma\left(\frac{\pi}{2},\frac{\pi}{4}\right)=\frac{8}{3}\left[\frac{100}{25+12(1-\cos\Delta\omega t)(1-P_{\rm H})}-1\right], \tag{10}$$

where $\gamma(\frac{\pi}{2},\frac{\pi}{4})=\frac{I_{\rm e}(\pi/2,t)}{I_{\rm e}(\pi/4,t)}$. Results are shown in Fig. 3. The solid (dashed) lines in different colors show the relations between γ and $P_{\rm H}$ for different time delays with (without) approximations used for $\Delta\omega_{ij}$ as mentioned before. Clearly the approximated results are sufficiently accurate. From Fig. 3 it is clear that we are able to determine $P_{\rm H}$ from γ if the time delay between the pump and probe pulses is carefully chosen, and it is most effective to choose the time delay to be half of the spin-orbit coupling time which is $t=0.5$ ns in this specific example. Note that γ changes from 1 to 8 as $P_{\rm H}$ changes from −1 to 1 if $t=0.5$ ns, which implies that we should be able to accurately determine $P_{\rm H}$ with a subnanosecond time resolution.

Another remark is that, by referring to Eq. (10), the results in Fig. 3 do not depend on the probe-pulse wavelength, since the matrix element R_{3d}^{kf}, which is generally wavelength dependent, does not appear in Eq. (10). We would like to emphasize once more that the proposed scheme is insensitive to the Doppler broadening of the target hydrogen gas as long as the pulse bandwidth exceeds the Doppler broadening, which is surely the case if we employ 1-ps pump and probe pulses. Hence a cooling technique is not required in our scheme to improve the detection efficiency [7].

The last interesting question is, "What is the relation between the degree of nuclear-spin polarization of hydrogen atoms $P_{\rm H}$ and that of produced protons upon photoionization ?" We have found that the relation between them strongly depends on the exact distribution of populations over the hyperfine ground states, and there is no single answer for that. However, it is clear that the spin polarization of produced protons upon photoionization is identical to $P_{\rm H}$ if $t=0$. This means that once we determine $P_{\rm H}$ through the procedure described above with an appropriate time delay, we can set it to zero so that spin polarization of the proton becomes identical with the measured value of $P_{\rm H}$.

B. Linearly and left-circularly polarized probe pulse

We also carry out similar calculations for the cases of linearly and left-circularly polarized probe pulses to find that the results are very similar to those for the case of right circularly polarized probe pulses. The only difference is that the results now depend on the atomic parameters such as the radial matrix elements, R_{3d}^{kp} and R_{3d}^{kf}, and the phase shifts, δ_p and δ_f, which means that the results are different for different wavelengths of the probe pulse. For instance, for the left-circularly polarized probe pulse, the expression for the PAD now reads

$$\begin{aligned}I_{\rm e}(\theta,t)\propto\ &\frac{8\pi}{625}f_3(\theta)(1-\cos\Delta\omega t)(1-P_{\rm H})\\&+\frac{\pi}{2500}f_4(\theta)(17+8\cos\Delta\omega t)(1-P_{\rm H})\\&+\frac{\pi}{100}f_4(\theta)(1+P_{\rm H}),\end{aligned} \tag{11}$$

where $f_3(\theta)$ and $f_4(\theta)$ are defined as

$$\begin{aligned}f_3(\theta)=\cos^2\theta\big[&4\big|R_{3d}^{kp}\big|^2+(2\cos^2\theta-3\sin^2\theta)^2\big|R_{3d}^{kf}\big|^2\\&+4(2\cos^2\theta-3\sin^2\theta)\cos(\delta_p-\delta_f)R_{3d}^{kp}R_{3d}^{kf}\big]\end{aligned} \tag{12}$$

and

$$\begin{aligned}f_4(\theta)=\sin^2\theta\big[&16\big|R_{3d}^{kp}\big|^2+(4\cos^2\theta-\sin^2\theta)^2\big|R_{3d}^{kf}\big|^2\\&+8(4\cos^2\theta-\sin^2\theta)\cos(\delta_p-\delta_f)R_{3d}^{kp}R_{3d}^{kf}\big].\end{aligned} \tag{13}$$

The three parameters R_{3d}^{kp}, δ_p, δ_f take the values of $R_{3d}^{kp}=0.57$ in atomic unit and $\delta_p=-2.30$, $\delta_f=2.22$ when the probe pulse has a wavelength of 410 nm. Similar to Eq. (10) for the right-circularly polarized probe pulse, the good choice of the detection angles are $\theta=0$ and $\pi/2$, and γ now reads

$$\gamma\left(0,\frac{\pi}{2}\right)=\frac{4f_3(0)}{f_4\left(\frac{\pi}{2}\right)}\left[\frac{1}{1-\frac{4}{25}(1-\cos\Delta\omega t)(1-P_{\rm H})}-1\right]. \tag{14}$$

Using Eq. (14) we calculate γ as a function of $P_{\rm H}$ and show the results in Fig. 4. It is clear that, similar to the case of

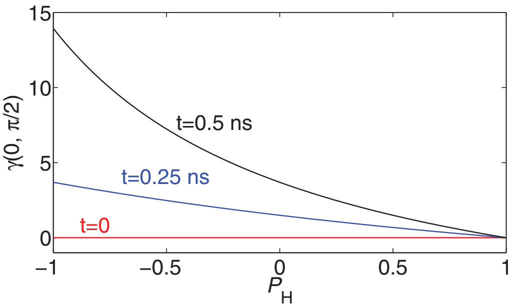

FIG. 4. (Color online) Similar to Fig. 3 but for the relation between $\gamma(0,\frac{\pi}{2})$ and $P_{\rm H}$ with the left-circularly polarized probe pulse.

right-circularly polarized probe pulses, we can determine P_{H} from the value of $\gamma(0,\pi/2)$.

III. ONE-PHOTON RESONANT TWO-PHOTON IONIZATION THROUGH THE $2p$ MANIFOLD

So far we have assumed to use the $3d$ manifold as the resonant intermediate states for the two-photon excitation. In principle, if a much shorter wavelength is available for the pump pulse, even a one-photon excitation can be used to reach the resonant intermediate states such as the $2p$ manifold. For instance, if both pump and probe pulses are right-circularly polarized, we obtain

$$\begin{aligned} I_{\mathrm{e}}(\theta,t) \propto\ & \frac{4\pi}{3} f_5(\theta)(1-\cos\Delta\omega' t)(1-P_{\mathrm{H}}) \\ & + \frac{\pi}{6} f_6(\theta)(5+4\cos\Delta\omega' t)(1-P_{\mathrm{H}}) \\ & + \frac{3\pi}{2} f_6(\theta)(1+P_{\mathrm{H}}), \end{aligned} \tag{15}$$

where $f_5(\theta)$ and $f_6(\theta)$ are defined as

$$f_5(\theta) = \sin^2\theta\cos^2\theta\left|R_{2p}^{kd}\right|^2 \tag{16}$$

and

$$f_6(\theta) = \sin^4\theta\left|R_{2p}^{kd}\right|^2, \tag{17}$$

where $\Delta\omega'$ is the fine-structure splitting between $2p\ ^2P_{1/2}$ and $^2P_{3/2}$, which is about 10 GHz, and R_{2p}^{kd} is the radial matrix element from the $2p$ state to the kd continuum state. The convenient angles to detect the photoelectron signals are $\theta = \pi/4$ and $\pi/2$, and γ reads

$$\gamma\left(\frac{\pi}{4},\frac{\pi}{2}\right) = \frac{1}{4} + \frac{1}{\frac{9}{(1-\cos\Delta\omega' t)(1-P_{\mathrm{H}})} - 2}. \tag{18}$$

The corresponding results are shown in Fig. 5. It is obvious that we can determine P_{H} from the value of $\gamma(\pi/4,\pi/2)$.

From the results we have presented in Secs. II A, II B, and III, it is clear that PADs via resonant excited states by multiphoton ionization always contain sufficient information on the initial degree of nuclear-spin polarization, which can be accurately determined only if laser polarization and detection angles of photoelectron signals are appropriately chosen.

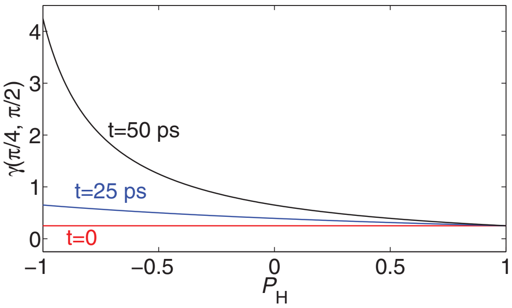

FIG. 5. (Color online) Similar to Fig. 3 but for the relation between $\gamma(\frac{\pi}{4},\frac{\pi}{2})$ and P_{H} when one-photon resonant two-photon ionization through the $2p$ manifold is employed. Both pump and probe pulses are assumed to be right-circularly polarized.

IV. CONCLUSIONS

In conclusion, we have theoretically demonstrated that the angle-resolved photoelectron signals produced by the pump and time-delayed probe pulses with short pulse durations can serve as a new type of nuclear-spin polarimeter. Specific results have been presented for hydrogen atoms using the fine-structure and hyperfine manifold of a $3d$ state as resonant intermediate states. Although we resolve neither fine-structure nor hyperfine states of the $3d$ manifold, we have clearly seen that the ratio of the photoelectron signals at two appropriately chosen angles and the time delay can be unambiguously correlated to the degree of nuclear-spin polarization. Because of the use of short laser pulses which is inherently broadband and the detection through photoelectron signals, the proposed scheme does not suffer from the Doppler broadening and works well under the low number density of atoms, such as an atomic beam, to be analyzed. The expected time resolution is as high as subnanosecond, and it is essentially limited by the spin-orbit coupling time of the resonant excited state. The present scheme is immediately applicable to other unstable elements such as $^{11}\mathrm{Be}^+$, $^{27}\mathrm{Mg}^+$, and $^{35}\mathrm{Ca}^+$, etc.

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