Effects of laser intensity and applied electric field on coherent control of spin polarization by short laser pulses

Takashi Nakajima[a)]

Institute of Advanced Energy, Kyoto University, Gokasho, Uji, Kyoto 611-0011, Japan and Institute for Solid State Physics (ISSP), The University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8581, Japan

We present a complete theory for coherent control of spin polarization through the pump-probe sequence, which is valid at arbitrary laser intensities. Anomalous intensity-dependent spin polarization has been found. Furthermore, as a new doorknob for coherent control, we propose an application of the electric field to effectively shorten the coupling time, which can be very effective for a nearly degenerate system. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2185260]

Rapid progress of ultrafast laser technology in recent years has enabled us to access to various interesting phenomena. In order to study ultrafast reactions, the use of a time-delayed pulse pair with a femtosecond or picosecond pulse duration is known to be one of the most powerful methods. Generally this is the so-called a pump-probe technique: the dynamical change created in matter by the pump pulse is investigated by the time-delayed probe pulse.

Alternatively, the pump-probe technique can be used for the purpose of active control (coherent control) of the photoabsorption dynamics not only in atoms/molecules[1,2] but also in semiconductors and quantum dots,[3–5] etc. Typically, the pump pulse coherently excites more than a single state. After the pump pulse, the system evolves in time in a free field, whose coupling time or evolution period depends on the energy interval between the coherently excited intermediate states. The time delay of the probe pulse with respect to the pump pulse is chosen in such a way to maximize the population of the target state. To apply the pump-probe technique for coherent control, however, the following issues have to be carefully dealt with: First, the intensities of the lasers are desired to be reasonably high so that maximum products are obtained, which is often well beyond the validity of transition-rate approximation. Second, the system we would like to apply the pump-probe technique may have (near)degeneracy in the intermediate states which are to be coherently excited by the pump pulse. If the intermediate states are (nearly)degenerate, the pump-probe technique would not be a good choice to control the interaction due to the very long coupling time during which the system may experience various kinds of relaxations such as the spontaneous/collisional decay, etc., before the arrival of the probe pulse. Thus, a new technique is desired to control not only the interaction itself but also the coupling time for a system with (near)degeneracy.

The aim of this letter is twofold. The primary aim is to present a complete theory for coherent control of spin polarization based on a pump-probe sequence, which is valid for arbitrary intensities and pulse durations. Related to this, the theoretical treatment reported in Refs. 6 and 7 is valid only in the weak intensities since the transition-rate approximation has been implicitly employed. The secondary aim is to propose an application of the electric field to control the

coupling time for a (nearly)degenerate system and discuss its effectiveness in a realistic context.

The level scheme we consider is shown in Fig. 1.[7] We excite Mg atoms in the ground $3s^2\ ^1S_0$ state to the triplet $3s3p\ ^3P_1(M_J=+1)$ state, labeled as $|0\rangle$, by a right-circularly polarized laser pulse (excitation pulse), with M_J being the projection of the total angular momentum J onto the quantization axis which is taken to be the propagation direction of the excitation pulse. Duration of the excitation pulse may be a few nanoseconds or longer, since its purpose is to prepare atoms in state $|0\rangle$, which will serve as an initial state for the subsequent pump-probe sequence. Once atoms are prepared in $|0\rangle$, we perform pump-probe photoionization by linearly polarized short laser pulses. As usual we assume that the polarization axis is parallel to the quantization axis. The pump pulse coherently excites the fine structure of $3s3d\ ^3D_1(M_J=+1)$ and $^3D_2(M_J=+1)$, which are labeled as $|1\rangle$ and $|2\rangle$, respectively. They are 0.031 cm^{-1} apart in energy without external fields. Note that we are dealing with a nearly degenerate system.

In order to describe the dynamics at arbitrary laser intensities and pulse durations, we derive a set of time-dependent amplitude equations which read

$$\dot{u}_0 = -i\Omega_1 u_1 - i\Omega_2 u_2, \tag{1}$$

$$\dot{u}_1 = \left[i\delta_1 - \tfrac{1}{2}(\gamma_1 + \gamma_1^{sp})\right]u_1 - i\Omega_1 u_0 - i\Omega_{12}^{(2)}u_2, \tag{2}$$
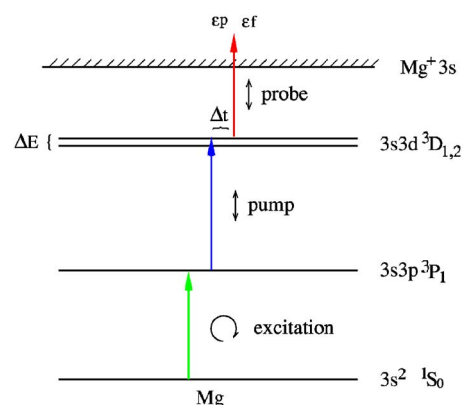

FIG. 1. (Color online) Level scheme.

[a)]Electronic mail: t-nakajima@iae.kyoto-u.ac.jp

$$\dot{u}_2 = \left[i\delta_2 - \tfrac{1}{2}(\gamma_2 + \gamma_2^{\rm sp}) \right] u_2 - i\Omega_2 u_0 - i\Omega_{12}^{(2)} u_1, \qquad (3)$$

where u_j $(j=0,1,2)$ are the probability amplitudes of states $|j\rangle$, and Ω_k $(k=1,2)$ are the Rabi frequencies between $|0\rangle$ and $|k\rangle$. δ_k are the detunings defined as $\delta_k = E_0 + \hbar\omega_{\rm pump} - (E_k + S_k)$ with E_k being the energy of $|j\rangle$, and S_k the shift if there is any additional external field. $\Omega_{12}^{(2)}$ is the complex two-photon Rabi frequency between $|1\rangle$ and $|2\rangle$ by the probe pulse. γ_j $(j=1,2)$ are the ionization widths of state $|j\rangle$ by the probe laser, while $\gamma_j^{\rm sp}$ represent the natural widths which is $(6.2\ {\rm ns})^{-1}$ for our specific case. Note that the values of $\Omega_{12}^{(2)}$ and γ_j $(j=1,2)$ depend on the probe photon energy. We emphasize that we do not necessarily have to assume[6,7] that the duration of the pump pulse, $\tau_{\rm pump}$, is sufficiently short compared to the coupling time, i.e., $\tau_{\rm pump} \ll (\Delta E)^{-1}$ where ΔE is defined as $\Delta E \equiv (E_1 + S_1) - (E_2 + S_2)$. Using the Hartree–Fock wave functions expanded on the discretized B-spline basis set,[8] we have calculated all the necessary atomic parameters such as Ωs, γs, and Ss, which have been obtained to be $\Omega_1 = -1.1 \times 10^8 \sqrt{I_{\rm pump}}$ (rad/s), $\Omega_2 = 1.48 \times 10^8 \sqrt{I_{\rm pump}}$ (rad/s), $\gamma_1 = 0.589\pi\left[\left(\tfrac{13}{150}\right)|R_{3d}^{\epsilon p}|^2 + \left(\tfrac{9}{50}\right)|R_{3d}^{\epsilon f}|^2 \right] I_{\rm probe}$ (s^{-1}), $\gamma_2 = 0.589\pi\left[\left(\tfrac{1}{6}\right)|R_{3d}^{\epsilon p}|^2 + \left(\tfrac{3}{14}\right)|R_{3d}^{\epsilon f}|^2 \right] I_{\rm probe}$ (s^{-1}), and ${\rm Im}\,\Omega^{(2)} = -0.589\pi/2\left[-(7\sqrt{5}/150)|R_{3d}^{\epsilon p}|^2 - (\sqrt{5}/50)|R_{3d}^{\epsilon f}|^2 \right] I_{\rm probe}$ (rad/s). $R_{3d}^{\epsilon l}$ $(l=p$ or $f)$ are the bound-free radial matrix elements (in atomic units) from $3s3d$ to $3s\epsilon l$ with ϵ being the continuum energy and $R_{3d}^{\epsilon p} = 51.2$ (1.51), and $R_{3d}^{\epsilon f} = -8.8$ (-26.7) for $\omega_{\rm probe} = 4.46$ (4.01) eV. Re $\Omega^{(2)}$ is $-9.8 I_{\rm probe}$ $(19.7 I_{\rm probe})$ (rad/s) for $\omega_{\rm probe} = 4.46$ (4.01) eV. Note that laser intensities $I_{\rm pump}$ and $I_{\rm probe}$ are given in W/cm^2.

Once the bound-state dynamics have been obtained by solving Eqs. (1)–(3), the partial photoelectron yields with specific spin can be obtained from the equations below:

$$R_e^{(\uparrow)} = 0.589\pi\left[\left| \tfrac{\sqrt{2}}{5\sqrt{3}}u_1 - \sqrt{\tfrac{2}{15}}u_2 \right|^2 |R_{3d}^{\epsilon p}|^2 + \left| \tfrac{3}{5\sqrt{14}}u_1 - \tfrac{3}{\sqrt{70}}u_2 \right|^2 \right.$$
$$\times |R_{3d}^{\epsilon f}|^2 + \left| -\tfrac{\sqrt{3}}{10}u_1 + \tfrac{1}{\sqrt{60}}u_2 \right|^2 |R_{3d}^{\epsilon p}|^2 + \left| -\tfrac{\sqrt{6}}{5\sqrt{7}}u_1 \right.$$
$$\left. + \sqrt{\tfrac{2}{105}}u_2 \right|^2 |R_{3d}^{\epsilon f}|^2 \right] I_{\rm probe}, \qquad (4)$$

$$R_e^{(\downarrow)} = 0.589\pi\left[\left| -\tfrac{\sqrt{3}}{10}u_1 + \tfrac{1}{\sqrt{60}}u_2 \right|^2 |R_{3d}^{\epsilon p}|^2 \right.$$
$$+ \left| -\tfrac{\sqrt{6}}{5\sqrt{7}}u_1 + \sqrt{\tfrac{2}{105}}u_2 \right|^2 |R_{3d}^{\epsilon f}|^2$$
$$\left. + \left| \sqrt{\tfrac{3}{35}}u_1 + \tfrac{1}{\sqrt{21}}u_2 \right|^2 |R_{3d}^{\epsilon f}|^2 \right] I_{\rm probe}, \qquad (5)$$

where the factors in front of $R_{3d}^{\epsilon l}$ are the angular coefficients for the relevant transitions. The temporal profiles of the pump and probe pulses are assumed to be Gaussian, i.e., $I_{\rm pump} = I_{\rm pump}^{(0)} \exp[-(t/\tau_{\rm pump})^2]$ and $I_{\rm probe} = I_{\rm probe}^{(0)} \exp\{-[(t - t_{\rm probe})/\tau_{\rm probe}]^2\}$ where $t_{\rm probe}$ is the delay of the probe pulse with respect to the pump pulse, and we assume the pulse durations to be $\tau_{\rm pump} = \tau_{\rm probe} = 10$ ps. In order to avoid the unnecessary Rabi oscillations, we further assume that the pump intensity is below the saturation intensity for this pulse duration, and set $I_{\rm pump}^{(0)} = 10^5$ W/cm^2.

Using Eqs. (4) and (5), we can calculate the spin polarization, $(R_e^{(\uparrow)} - R_e^{(\downarrow)})/(R_e^{(\uparrow)} + R_e^{(\downarrow)})$, for different photon energies and intensities of the probe laser. Figure 2(a) shows the variation of spin polarization as a function of delay time, $t_{\rm probe}$, for four different probe laser intensities, $I_{\rm probe}^{(0)} = 10^7$, 10^8, 10^9, and 10^{10} W/cm^2 with $\omega_{\rm probe} = 4.46$ eV. It is very interesting to point out that spin polarization exhibits anoma-
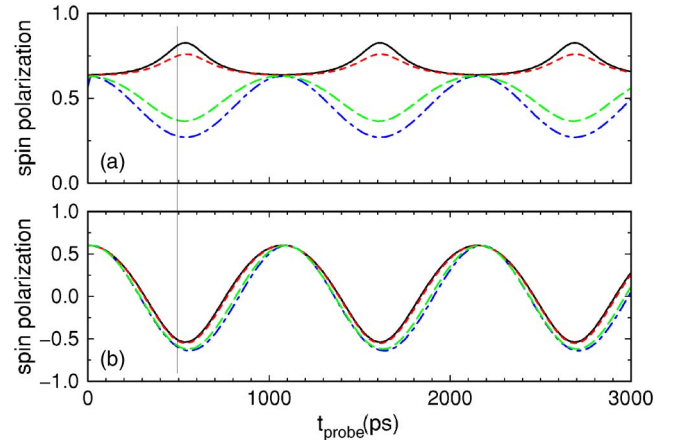

FIG. 2. (Color online) Spin polarization as a function of time delay, $t_{\rm probe}$, for (a) $\omega_{\rm probe} = 4.46$ eV and (b) $\omega_{\rm probe} = 4.01$ eV. In both panels $I_{\rm probe}^{(0)} = 10^7$ W/cm^2 (solid black), 10^8 W/cm^2 (dashed red), 10^9 W/cm^2 (long-dashed green), and 10^{10} W/cm^2 (dot-dashed blue).

lous dependence on $I_{\rm probe}^{(0)}$. On the contrary, spin polarization has very little dependence on $I_{\rm probe}^{(0)}$ for $\omega_{\rm probe} = 4.01$ eV, as shown in Fig. 2(b). In order to understand why spin polarization in Figs. 2(a) and 2(b) is so different for the different probe photon energies, we look into Eqs. (4) and (5) more carefully, where we find that the only parameters that depend on $I_{\rm probe}$ are u_1 and u_2, since $R_{3d}^{\epsilon p}$ and $R_{3d}^{\epsilon f}$ depend only on $\omega_{\rm probe}$. We have calculated the temporal evolution of u_1 and u_2 at the two different probe intensities, $I_{\rm probe}^{(0)} = 10^7$ and 10^9 W/cm^2, for the delay of $t_{\rm probe} = 500$ ps, corresponding to the time delay given by the vertical line in Figs. 2(a) and 2(b). The results are shown in Fig. 3 for $\omega_{\rm probe} = 4.46$ eV [Figs. 3(a)–3(d)] and 4.01 eV [Figs. 3(e)–3(h)], respectively. At the time of 500 ps, depletion of the probability amplitudes is seen in all graphs due to ionization by the probe laser. The amount of depletion, however, is very different for different $\omega_{\rm probe}$ and $I_{\rm probe}^{(0)}$, which can be attributed to the different ionization widths for different $\omega_{\rm probe}$ and $I_{\rm probe}^{(0)}$. In particular, depletion of u_2 is significant when $\omega_{\rm probe} = 4.46$ eV and

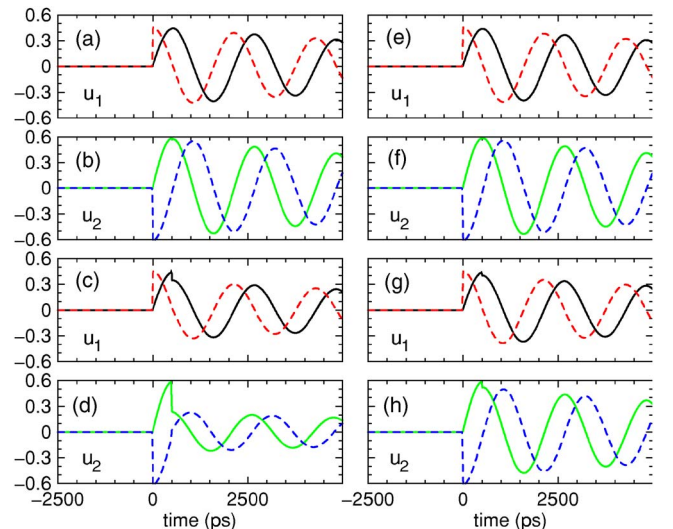FIG. 3. (Color online) Time evolution of u_1 and u_2 at $t_{\rm probe} = 500$ ps. (a) and (b) $\omega_{\rm probe} = 4.46$ eV at $I_{\rm probe}^{(0)} = 10^7$ W/cm^2, and (c) and (d) $I_{\rm probe}^{(0)} = 10^9$ W/cm^2. (e) and (f) $\omega_{\rm probe} = 4.01$ eV at $I_{\rm probe}^{(0)} = 10^7$ W/cm^2, and (g) and (h) $I_{\rm probe}^{(0)} = 10^9$ W/cm^2. In each graph, solid and dashed lines represent the real and imaginary parts of the probability amplitude, respectively.
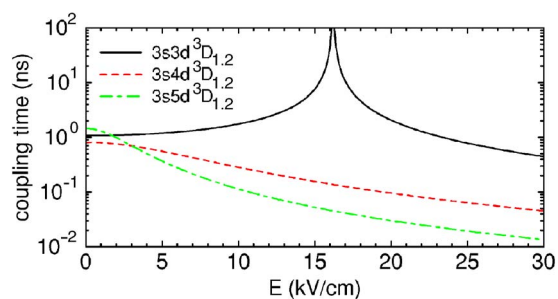
FIG. 4. (Color online) Variation of the effective coupling time as a function of electric field strength. Electric field is assumed to be parallel to the laser polarization axis.

$I_{\text{probe}}^{(0)} = 10^9$ W/cm^2 [Fig. 3(d)]. Since the photoelectron yield into each spin state critically depends on u_1 and u_2 multiplied by the pulse function, I_{probe}, as shown in Eqs. (4) and (5), we now understand why spin polarization in Figs. 2(a) and 2(b) behaves so differently for different probe photon energies.

So far we have assumed that there is no static electric field, i.e., $S_1 = S_2 = 0$. Now we apply the static electric field and consider how the system behavior changes. Needless to say, the application of the static electric field induces dc stark shifts. Since dc stark shifts are state dependent, the energy difference between $|1\rangle$ and $|2\rangle$ will be effectively modified, which leads to the change of the coupling time. With the bound-free as well as bound-bound transition moments obtained from the Hartree–Fock code, we have calculated the dc stark shifts for $3snd\,^3D_{1,2}$ ($n = 3, 4, 5$) with which the effective coupling time is computed for a given electric field strength. Figure 4 shows the variation of the effective cou-

pling time as a function of applied electric field strength. We see that the behavior is different for different n simply because dc stark shifts are state dependent. Figure 4 clearly demonstrates that the application of the external electric field can be a new and effective doorknob for a system with (near)degeneracy to control the coupling time for a pump-probe sequence.

In summary, we have developed a complete theory for coherent control of spin polarization through the pump-probe sequence. The developed theory works at arbitrary laser intensities and pulse durations. Anomalous intensity-dependent spin polarization has been found. We have also proposed the use of a static electric field to control the coupling time through state-dependent dc stark shifts, which can be a new doorknob to control the coupling time.

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