The time-scale of nonlinear events driven by strong fields: Can one control the spin-coupling before ionization runs over?

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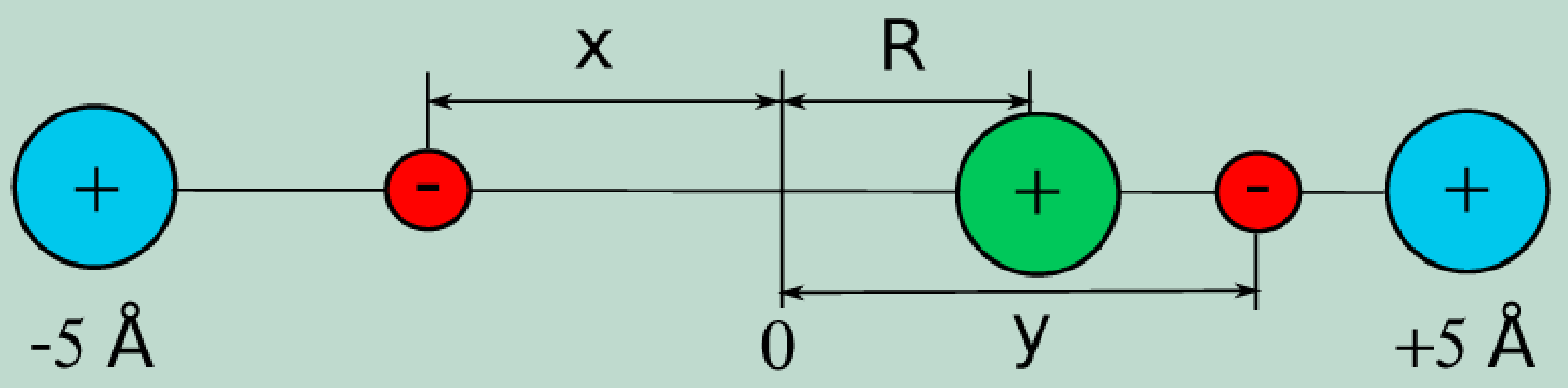
Introduction

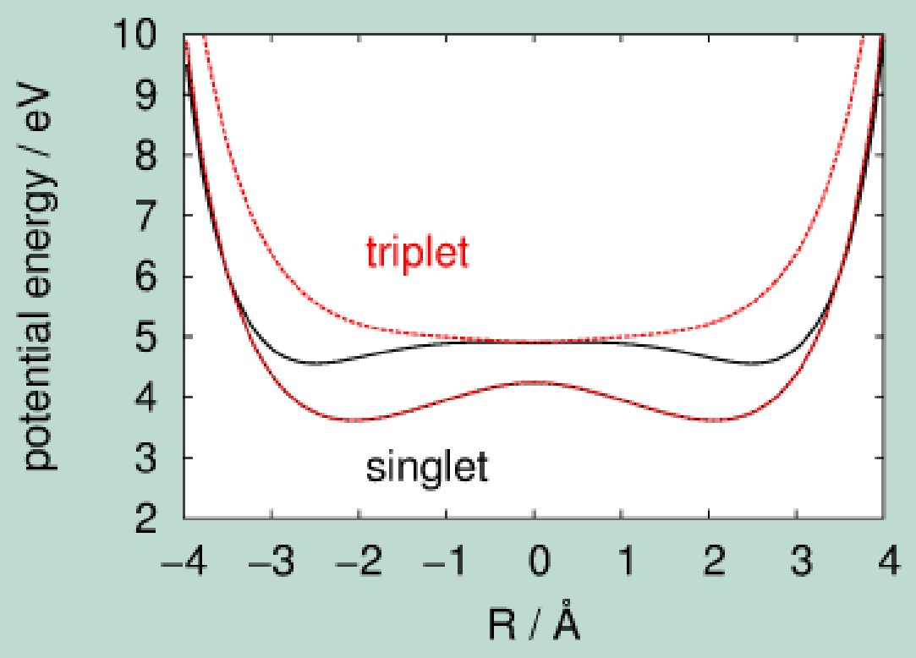
The non-resonant dynamic Stark effect (NRDSE) can be used for the optical control of the spin state of a system. The electric field applied to generate the Stark effect can also lead to ionization. In our work, we investigate the relation between spin coupling and ionization rate using a simple two-level Hamiltonian and identify conditions for an efficient spin-control by suppressing ionization.



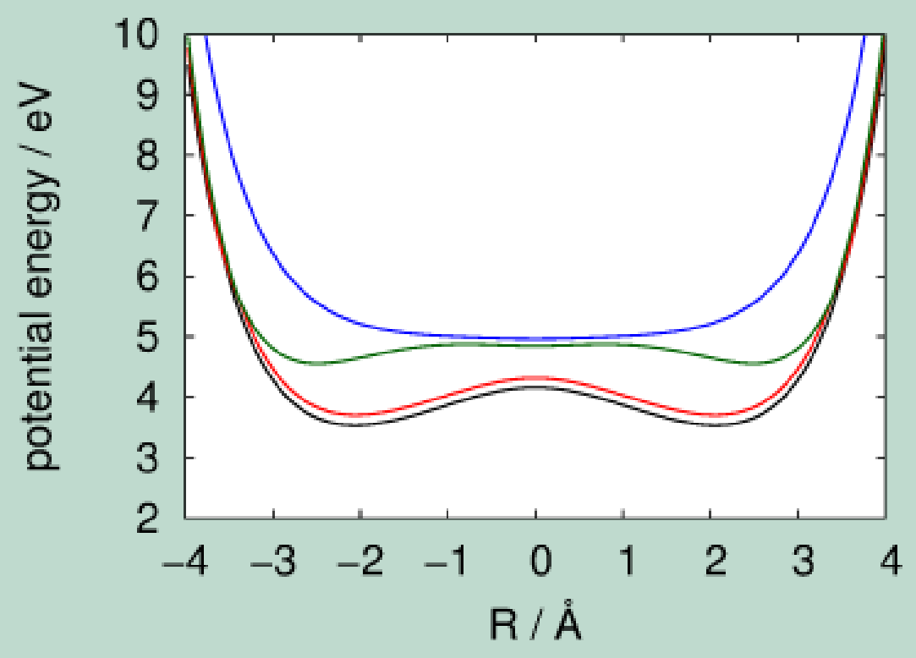
The results are confirmed in solving the time-dependent Schrödinger equation for the interaction of a laser field with a spin-coupled model system where two electrons and a nucleus move in a collinear configuration. We show that quantum control of intersystem crossing can indeed be effective if the intensity of the external field and the accompanying Stark shift are adjusted properly to the spin coupling-strength [1].

Field free spin dynamics

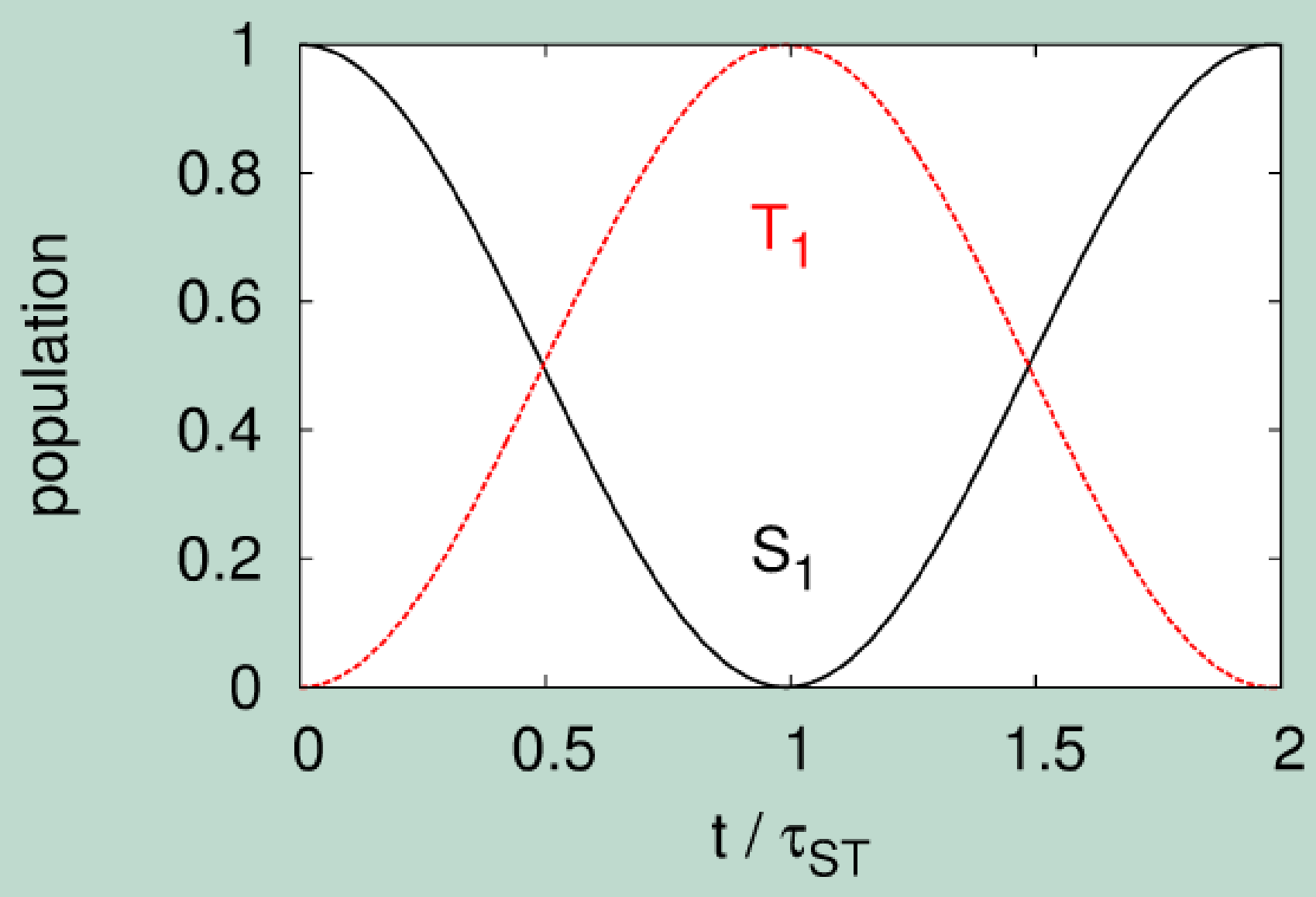
Solution of the electronic Schödinger equation yields potential energy curves [2, 3]



$$\xrightarrow{\text{spin coupling}} \begin{pmatrix} H(x,y,R) & \lambda(x-y) \\ \lambda(x-y) & H(x,y,R) \end{pmatrix}$$



- groundstate singlet S_1 and triplet T_1 nearly degenerate
- spin coupling causes level repulsion



initial population in S_1
↓
spin transition dynamics $S_1 \to T_1$
with characteristic time $\tau_{ST} = \frac{\pi}{2V_{ST}}$
depending on singlet-triplet coupling V_{ST}
↓
triplet population:

$$P_{T_1}(t) = \sin^2(V_{ST} \cdot t)$$

Model consideration: Control of spin transfer via NRDSE

Simple model of spin dynamics: two-level Hamiltonian only including S_1 and T_1

$$\mathrm{H}_{\mathrm{eff}} = \begin{pmatrix} -i\Gamma(E) & V_{ST} \\ V_{ST} & \Delta(E) - i\Gamma(E) \end{pmatrix}$$

Stark shift is

$$\Delta(E) = \Delta(0) - (\alpha_T - \alpha_S)E^2/2$$

in our case: $\Delta(0) = 0$

triplet population:

$$P_T(t) = e^{-\Gamma t}\left(\frac{V_{ST}}{\Omega_e}\right)^2 \sin^2 \Omega_e t$$

with frequency $\Omega_e = \sqrt{V_{ST}^2 + \Delta(E)^2}$ and ionization rate Γ

The NRDSE gives a prescription to avoid the spin transfer: Use a field sufficiently strong so that $\Delta(E) \gg V_{ST}$.

two timesacles:

- τ_{ST} for spin transfer: $\tau_{ST} = \frac{\pi}{2V_{ST}}$
- τ_{ion} for ionization: $\tau_{ion} = \frac{\ln 2}{\Gamma}$

$\Rightarrow$ spin locking possible if $\tau_{ion} \leq \tau_{ST}$

Adjusting spin coupling strength

maximum threshold value for the triplet population:

$$P_T^{max} = \frac{V_{ST}^2}{V_{ST}^2 + \Delta_m^2}$$

$$\Delta_m = V_{ST}\sqrt{\frac{1 - P_T^m}{P_T^m}} = V_{ST}\sqrt{\frac{P_S^{min}}{P_T^{max}}} \Rightarrow E^2 = \frac{2V_{ST}\chi^{1/2}}{\alpha_T - \alpha_S}$$

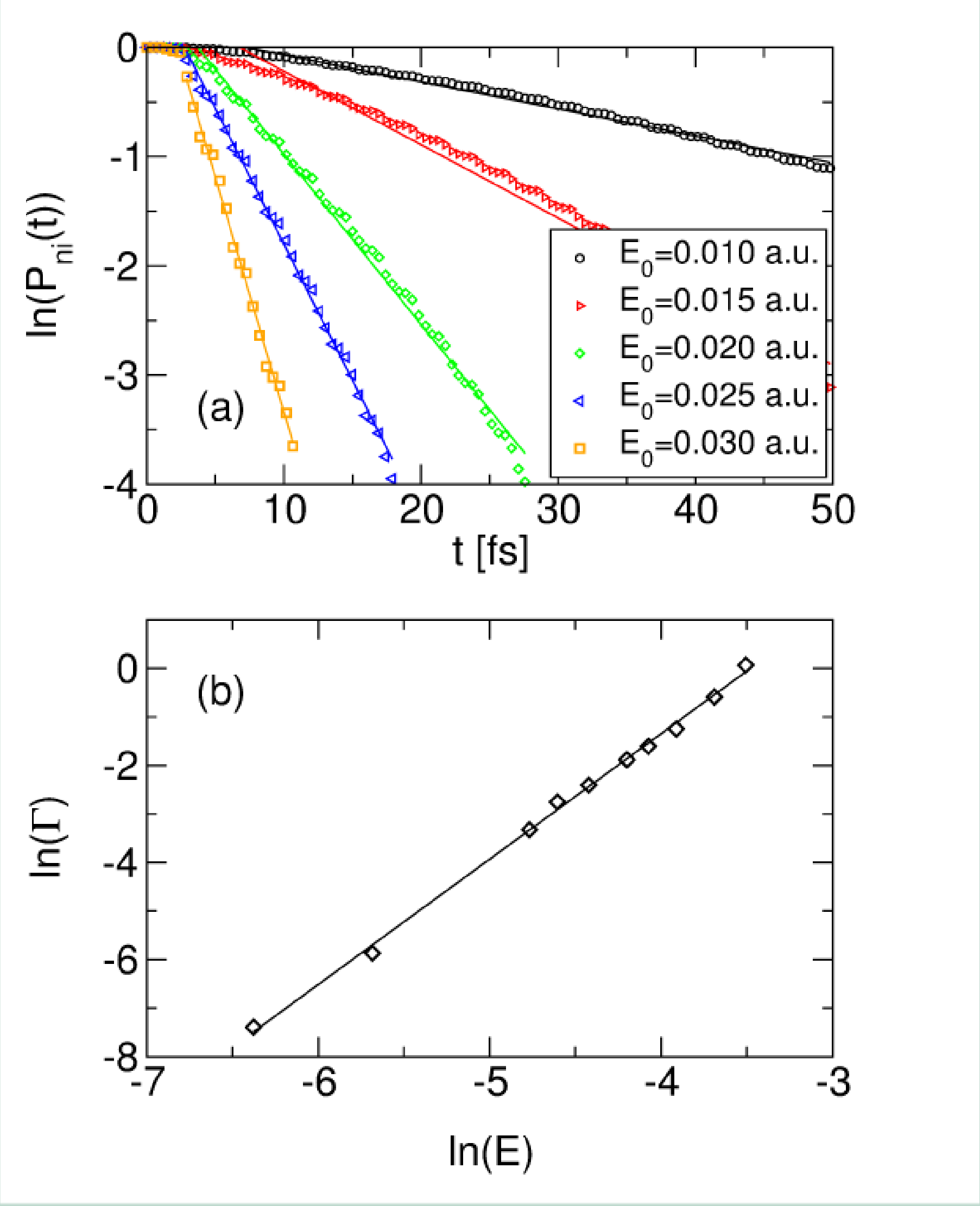
for efficient control:

$$\frac{\tau_{ST}}{\tau_{ion}} = K\left(\frac{P_S^{min}}{P_T^{max}}\right)^{n/4} V_{ST}^{(n-2)/2}$$

- $n = 2$: timescales fixed
- $n \neq 2$: control of spin transfer possible for appropriate values of V_{ST}

aim: $\tau_{ion} \approx \tau_{ST}$ and large $\frac{P_S^{min}}{P_T^{max}}$

Timescale of ionization



Ionization behaviour of the system calculated from numerically exact simulation:

- estimate ionization rate Γ from non-ionized part $P_{ni}(t)$:
  $$\ln(P_{ni}) \approx -\Gamma t$$
- simple approximation for ionization behaviour:
  $$\Gamma = CE^n$$
  $n/2$: effective photon number required to reach ionization

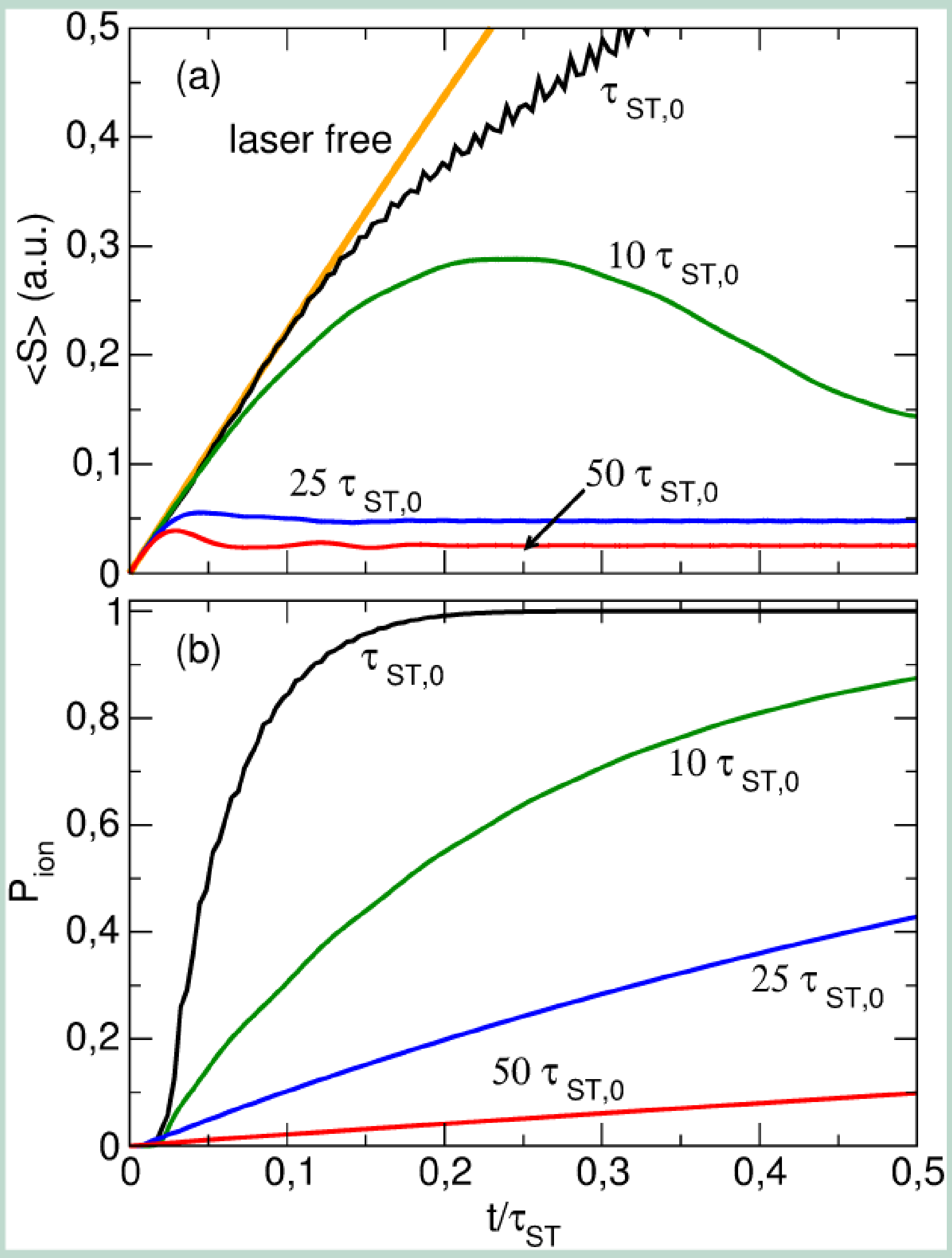
linear fit of $\ln E$ vs. $\ln \Gamma$
↓
effective photon number $n = 2.6$
⇓
spin locking possible

Spin control before onset of ionization

Starting parameters for control:
$E_0 = 0.017$ a.u. for $\lambda_0 = 1.028 \cdot 10^{-3}$ eV/Å

→ $\tau_{ST,0} = 120$ fs $\gg \tau_{ion,0} \approx 22$ fs
→ significantly weaker coupling neccessary

$$\frac{V_{ST}}{V_{ST,0}} = \left(\frac{\tau_{ST}}{\tau_{ion}}\frac{\tau_{ion,0}}{\tau_{ST,0}}\right)^{2/(n-2)} \approx \left(\frac{\tau_{ion,0}}{\tau_{ST,0}}\right)^{2/(n-2)} \approx \left(\frac{1}{6}\right)^{2/(n-2)} \approx \frac{1}{390}$$



observables:

- average spin angular momentum S_{av}
- probability of ionization P_{ion}

parameters:

- spin coupling strength $V_{ST,0}/m$ for $m = \{1, 10, 25, 50\}$ with corresponding spin transfer times $m \cdot \tau_{ST,0}$
- electric field strength $E_0/\sqrt{m}$ needed for control

results:

→ ionization still dominates for $V_{ST,0}$ and $V_{ST,0}/10$
→ for weaker couplings spin locking can be achieved before significant ionization occurs

Conclusion

We show that spin locking with the help of external electric fields can be achieved under certain conditions. To identify these conditions in our model system, we analyse the timescales of ionization and control of spin transfer via the nonresonant dynamic Stark effect (NRDSE) scheme in a simple two-level approximation of the system. Wether optical spin control is possible or not is mainly determined by the dependence of the ionization rate on the control field amplitude. As a simple empirical model we approximate this dependence as a power law whose exponent decides if ionization or control is the predominant process. This gives an estimate for the approptiate strength of spin coupling and control field which enable the control of the spin transfer. For relatively weak spin couplings and control field intensities we achieve efficient spin locking in an initial singlet state.

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