Nonlinear dichroism in double ionization of He by an intense elliptically-polarized few-cycle XUV pulse

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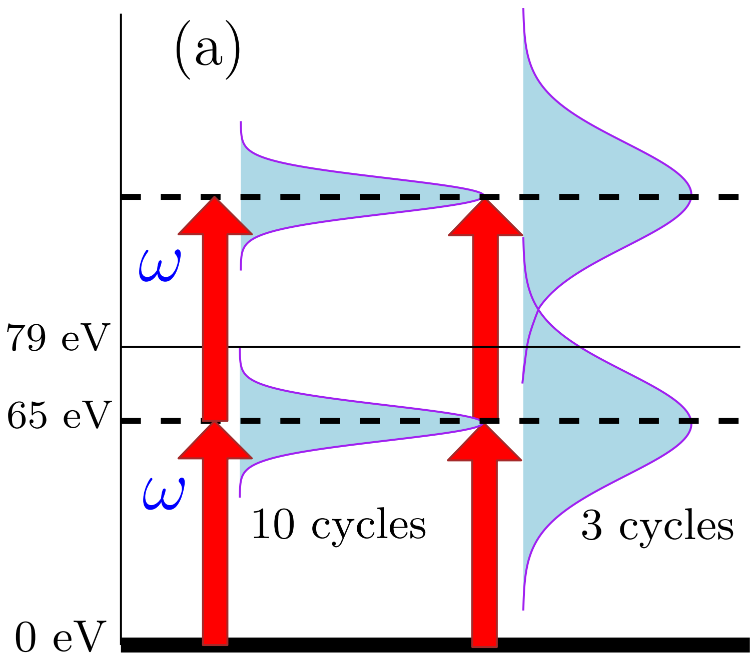
Short Pulse vs Long Pulse Double Ionization (DPI) of He



- **Pulse Parameters**
  - $\hbar\omega = 65$ eV
  - $\Delta\omega \simeq 1.44\omega/n$
  - $\Delta\omega = 31.2$ eV for $n = 3$
  - $T = 191$ as (3 cycles)
  - $I = 1 - 2 \times 10^{15}$ W/cm^2

Outline

1. Perturbation theory for double ionization of He (^{1}S^e) by an arbitrarily-polarized short pulse
2. Six-dimensional two-electron TDSE in the presence of an arbitrarily-polarized short pulse
3. TDSE results for the “in-plane” geometry (i.e., the two electron momenta lie in the polarization plane). We examine the sensitivity of the nonlinear dichroism (ND) to:
   - CEP
   - Intensity
   - Ellipticity
   - Energy-sharing
4. Conclusions

1. Perturbation theory for double ionization of He ($^1S^e$) by an arbitrarily-polarized short pulse

Perturbation Theory (PT) Formulation

- Triply differential probability (TDP):
  $d^3W/dEd\Omega_{\hat{\mathbf{p}}_1}d\Omega_{\hat{\mathbf{p}}_2} \equiv \mathcal{W}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{e}) = \mathcal{C}|A_1 + A_2|^2$

- The TDP for negligible $|A_2|^2$:
  $\mathcal{W}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{e}) \approx \mathcal{C}\left[|A_1|^2 + 2\mathrm{Re}\left(A_1^* A_2\right)\right]$

- *Ref*: J.M. Ngoko Djiokap *et al.*, *Phys. Rev. A* **88**, 053411 (2013)

- Absolute Dichroism:
  $\Delta\mathcal{W}_\xi = \mathcal{W}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{e}) - \mathcal{W}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{e}^*) \simeq \Delta\mathcal{W}_{D1} + \Delta\mathcal{W}_{D12}$

- Electric field: $\boldsymbol{F}(t) = \frac{1}{2}F_0(t)[\mathbf{e}e^{-i(\omega t+\phi)} + \mathbf{e}^* e^{i(\omega t+\phi)}]$

- Pulse polarization vector: $\mathbf{e} = (\hat{\epsilon} + i\eta\hat{\zeta})/\sqrt{1+\eta^2}$

PT parameterization of A_1 and A_2

- Reduced electron momenta: $\mathbf{p}_\pm = (\hat{\mathbf{p}}_1 \pm \hat{\mathbf{p}}_2)/2$.

- 1st-order amplitude for one-photon absorption:

  $A_1 = e^{-i\phi}[f_g(\rho)(\mathbf{e}\cdot\hat{\mathbf{p}}_+) + f_u(\rho)(\mathbf{e}\cdot\hat{\mathbf{p}}_-)]$

  - $f_g(p_2, p_1, \theta) = f_g(p_1, p_2, \theta)$
  - $f_u(p_2, p_1, \theta) = -f_u(p_1, p_2, \theta)$
  - EES + BTB: $A_1 = 0$ because $f_u = 0$ and $\mathbf{p}_+ = 0$.

- 2nd-order amplitude:

  $A_2 = e^{-2i\phi}B(\rho; \hat{\mathbf{p}}_+, \hat{\mathbf{p}}_-, \mathbf{e}, \mathbf{e}) + B'(\rho; \hat{\mathbf{p}}_+, \hat{\mathbf{p}}_-, \mathbf{e}, \mathbf{e}^*)$

  - Two-photon absorption:

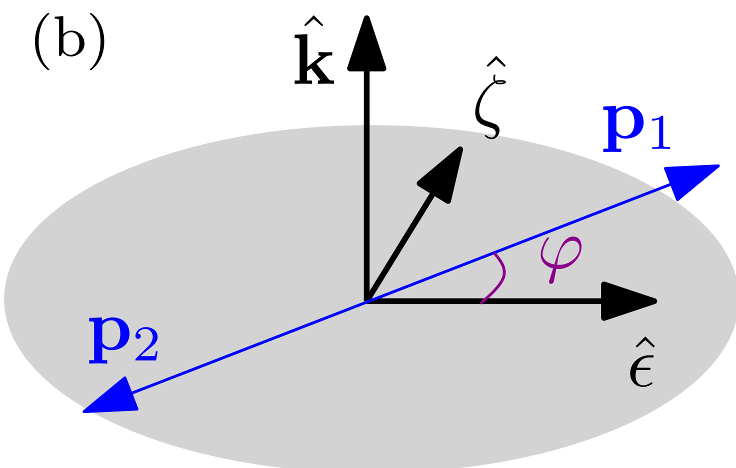
    $B = h(\mathbf{e}\cdot\mathbf{e}) + h_+(\hat{\mathbf{p}}_+\cdot\mathbf{e})^2 + h_-(\hat{\mathbf{p}}_-\cdot\mathbf{e})^2 + h_0(\hat{\mathbf{p}}_+\cdot\mathbf{e})(\hat{\mathbf{p}}_-\cdot\mathbf{e})$

  - One-photon absorption & one-photon emission (vice versa):

    $B' = h' + h'_+|\hat{\mathbf{p}}_+\cdot\mathbf{e}|^2 + h'_-|\hat{\mathbf{p}}_-\cdot\mathbf{e}|^2 + h'_0\,\mathrm{Re}\,\{(\hat{\mathbf{p}}_+\cdot\mathbf{e})(\hat{\mathbf{p}}_-\cdot\mathbf{e}^*)\}$

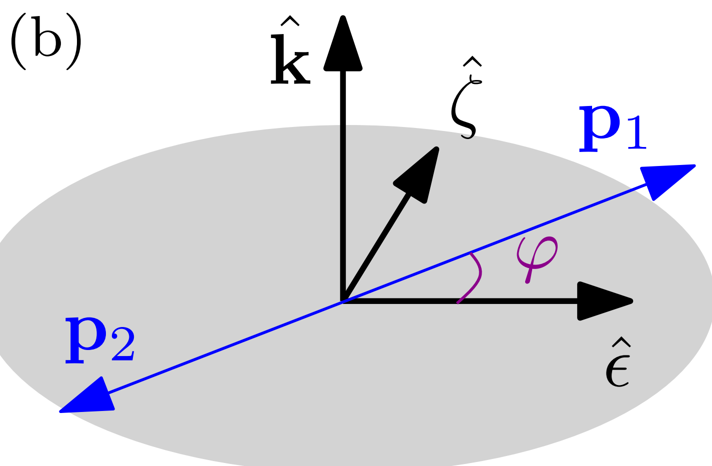
First-order dichroism from PT

- Absolute dichroism: $\Delta\mathcal{W}_\xi = \Delta\mathcal{W}_{D1} + \Delta\mathcal{W}_{D12}$
- Absolute first-order dichroism:
  $\frac{1}{C}\Delta\mathcal{W}_{D1} = |A_1(\mathbf{e})|^2 - |A_1(\mathbf{e}^*)|^2 = \pm\xi \sin\theta\, \mathrm{Im}[f_g^* f_u]$
  - Dependent on ξ
  - Independent of the CEP ϕ and the angle φ
  - $\Delta\mathcal{W}_{D1} \propto I^1$
  - UES+BTB geometry: $\theta = \pi$ and $\Delta\mathcal{W}_{D1} = 0$.



Nonlinear dichroism (ND) from PT

- Absolute dichroism: $\Delta\mathcal{W}_{\xi} = \Delta\mathcal{W}_{D1} + \Delta\mathcal{W}_{D12}$
- Absolute nonlinear dichroism for UES + BTB:
  $\Delta\mathcal{W}_{D12} = \mathcal{C}\xi\sqrt{2/(\ell+1)}\sin\varphi\,\mathrm{Im}\{f_u^*[e^{-i\phi}(2h\ell - h_-(\ell\cos 2\varphi + 1)) + e^{i\phi}(2h' + h'_-(\ell\cos 2\varphi + 1))]\}$
  - Dependent on ξ and $\xi\ell$
  - Dependence on both the CEP ϕ and φ
  - $\Delta\mathcal{W}_{D12} \propto I^{3/2}$, a key finding as $2\mathrm{Re}\,(A_1^* A_2)$



2. Rotating frame for solving the six-dimensional two-electron TDSE in presence of an arbitrarily-polarized few-cycle XUV pulse

The electric field (using the length form) seen by an observer in the rotating frame is always linearly-polarized.

Two-electron TDSE for an arbitrarily-polarized few-cycle pulse

- 6-dimensional TDSE: $i\partial_t \Phi(\mathbf{r}_1, \mathbf{r}_2, t) = H(t)\Phi(\mathbf{r}_1, \mathbf{r}_2, t)$
  - Hamiltonian: $H \equiv T_1 + T_2 + V_A + V_L(t)$
  - Atomic potential:
    $V_A = V_C + V_{12} \equiv -Z/r_1 - Z/r_2 + 1/r_{12}$
  - Laser-electron interaction: $V_L(t) = \mathbf{F}(t) \cdot (\mathbf{r}_1 + \mathbf{r}_2)$
- Pulse parameterization: $\mathbf{F}(t) = -\partial_t \mathbf{A}(t)$ and
  $\mathbf{A}(t) = \frac{(-1)^n A_0 f(t)}{\sqrt{1+\eta^2}}[\sin(\omega t + \phi)\hat{\epsilon} - \eta \cos(\omega t + \phi)\hat{\zeta}]$
- Close-coupling expansion:
  $\Phi(\mathbf{r}_1, \mathbf{r}_2, t) = \sum_{LM} \sum_{l_1 l_2} \frac{\Psi^{LM}_{l_1 l_2}(r_1, r_2, t)}{r_1 r_2} \Lambda^{LM}_{l_1, l_2}(\hat{r}_1, \hat{r}_2)$
- Electron angular momenta: $(-1)^L = (-1)^{l_1 + l_2}$
- $L = 0 - 3$ and $-L \leq M \leq +L$, $l_1 = l_2 = 0 - 5$

M-mixing problem

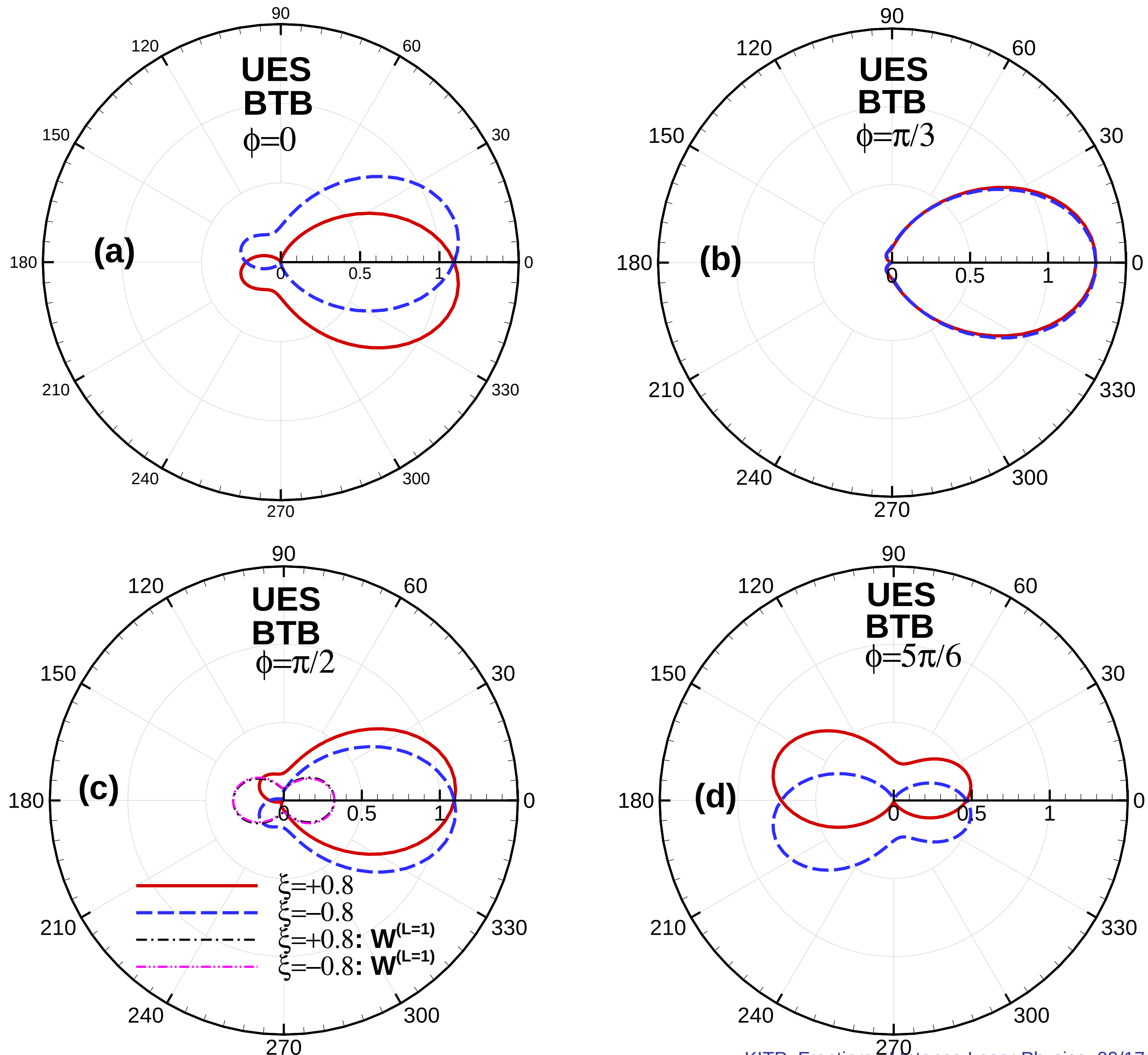
- For pulses with $\eta \neq 0$, $[H, L_z] \neq 0$, M is not conserved.
  - Basic ideas and principles of the method:
    - H.G. Muller, Laser Physics **9**, 138 (1999)
    - T. K. Kjeldsen *et al.*, Phys. Rev. A **75**, 063427 (2007)
  - Atomic interaction is treated in the atomic fixed frame in the laboratory coordinate system
  - Laser interaction is treated in the rotating frame defined by the external polarization vector.
  - Split-operator + rotation transformation (accuracy$\sim \tau^3$):
    $\Phi(t+\tau) =$
    $e^{-i\frac{\tau}{2}T} e^{-i\frac{\tau}{2}V_C} e^{-i\frac{\tau}{2}V_{12}} D^\dagger e^{-i\tau V_L} D e^{-i\frac{\tau}{2}V_{12}} e^{-i\frac{\tau}{2}V_C} e^{-i\frac{\tau}{2}T} \Phi(t)$

Relevant physical observable

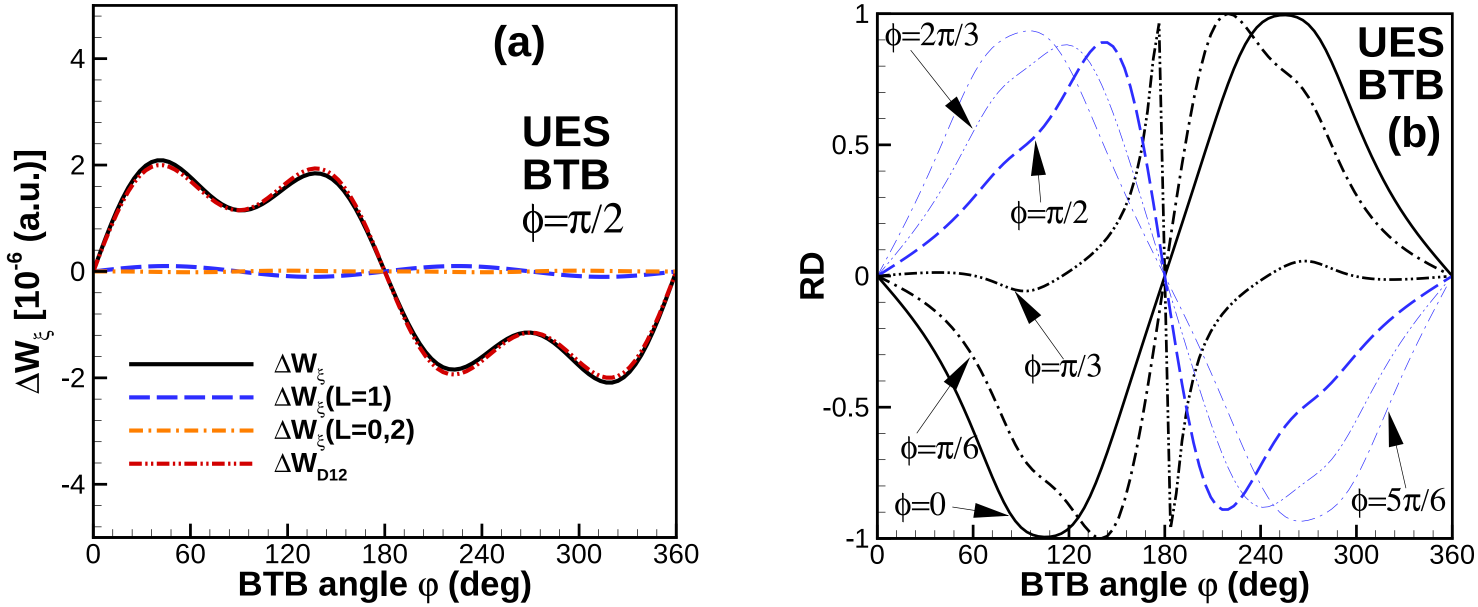
- The TDP calculated numerically:
  $\mathcal{W}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{e}) = |\langle \Theta^{(-)}_{\mathbf{p}_1,\mathbf{p}_2}(\boldsymbol{r}_1, \boldsymbol{r}_2)|\Phi_C(\boldsymbol{r}_1, \boldsymbol{r}_2, T + T', \phi, \mathbf{e})\rangle|^2$
  - Field-free double continuum states: $\Theta^{(-)}_{\mathbf{p}_1,\mathbf{p}_2}(\boldsymbol{r}_1, \boldsymbol{r}_2)$
  - Φ_C is the continuum part of the wave packet obtained T' after the pulse.
  - We have determined T' for convergence
  - Unequal energy sharing (UES): $E_1 = 0.7$ eV and $E_2 = 3.3$ eV
  - BTB scheme guarantees a high accuracy of our numerical method in the XUV regime [see M. Gailitis, J. Phys. B **23**, 85 (1990)]

3. Numerical results and analysis using our PT

Angular distributions for $\xi = \pm 0.8$ vs. the pulse CEP at 2 PW/cm^2



Dichroism vs. CEP



- $\Delta\mathcal{W}_{D1} \approx 0$, consistent with PT
- The significance of $\Delta\mathcal{W}_{D2}$ depends on the CEP
- At a CEP $\phi = \pi/2$, $\Delta\mathcal{W}_{\xi} \simeq \Delta\mathcal{W}_{D12}$ as predicted by PT
- PT predicts $\Delta\mathcal{W}_{D12} \propto \sin(\varphi)$, $\Delta\mathcal{W}_{D12}(\varphi = 0, \pi) = 0$
- $RD = \Delta\mathcal{W}_{\xi}/[\mathcal{W}(\mathbf{p}, \mathbf{e}) + \mathcal{W}(\mathbf{p}, \mathbf{e}^{*})]$ is very sensitive to the CEP; for nearly all CEPs, it is substantial.

Conclusions

- A new type of dichroic effect (the nonlinear dichroism) is predicted by our PT and demonstrated by our six-dimensional two-electron TDSE calculations.
- The nonlinear dichroism (ND) originates from the interference between the first- and the second-order PT amplitudes, owing to the broad bandwidth of a few-cycle pulse.
- We found that ND is sensitive to the CEP, ellipticity, intensity, and energy-sharing configuration.
- ND probes electron correlation on their natural timescale, since ND vanishes for long pulse.
- When intense attosecond pulses become a reality, reaction microscope techniques will demonstrate the ND.

Acknowledgments

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