# Modulation of Attosecond Beating in Resonant Two-Photon Ionization

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# Outline



- Outline of the resonant two-photon finitepulse model
- Extension to the multichannel case and comparison with experiment in argon



# **High Harmonic Generation**



T. Popmintchev et al., Nature Photonics 4, 822 (2010)

# **Attosecond Pulse Trains**



P. M. Paul et al., Science 292, 1689 (2001)

# Reconstruction of Attosecond Beating By Two photon transitions (RABBIT)



$$A + Y_{Hn} \rightarrow A^+ + e^-$$



K. Klünder et al., PRL 106, 143002 (2011)

 $A + Y_{Hn} \pm Y_{IR} \rightarrow A^+ + e^-$ 



# The principle of RABBIT

$$\Delta \mathcal{W}_{2n\omega}^{(2)} \propto |\mathcal{M}_{2n-1}\mathcal{M}_{2n+1}| \cos\left(2\omega_{_{\mathrm{IR}}}\tau - \Delta\phi_{2n} - \Delta\varphi_{2n}^{At}\right)|$$

 $\mathcal{M}_{2n\pm 1} = \langle E | \mathcal{O} \left[ G_0^+(\omega_g \mp \omega_{\mathrm{IR}}) + G_0^+(\omega_g + \omega_{2n\pm 1}) \right] \mathcal{O} | g \rangle$ 



# What if one paths is resonant?



$$\begin{array}{l} \left\{ \begin{array}{l} \mathsf{A} + \Upsilon_{2n-1} \rightarrow \mathsf{A}^{*}\left(\mathbf{r}\right) \\ \mathsf{A}^{*}(\mathbf{r}) + \Upsilon_{1\mathbb{R}} \rightarrow \mathsf{A}^{+} + \mathbf{e}^{-} \\ \mathsf{A} + \Upsilon_{2n+1} \rightarrow \mathsf{A}^{+} + \mathbf{e}^{-} + \Upsilon_{1\mathbb{R}} \\ \end{array} \right. \\ \left. \mathcal{M}_{2n-1} \simeq \left\langle E | \mathcal{O} G_{0}^{+}(\omega_{g} + \omega_{2n-1}) \mathcal{O} | g \right\rangle \\ \simeq \frac{\mathcal{O}_{Er} \mathcal{O}_{rg}}{\delta \omega + i0^{+}} \qquad \varphi_{n}^{At} = \arg \mathcal{M}_{n} \\ \end{array} \\ \left. \begin{array}{c} \mathcal{O}_{Er} \mathcal{O}_{rg} \\ \varphi_{n}^{At} = \arg \mathcal{M}_{n} \\ \end{array} \right. \\ \left. \begin{array}{c} \mathcal{O}_{er} \mathcal{O}_{rg} \\ \mathcal{O}_{er} \mathcal{O}_{er} \mathcal{O}_{er} \\ \mathcal{O}_{er} \mathcal{O}_{er} \mathcal{O}_{er} \\ \mathcal{O}_{er} \mathcal{O}_{er} \mathcal{O}_{er} \\ \end{array} \right. \\ \left. \begin{array}{c} \mathcal{O}_{er} \mathcal{O}_{er} \\ \mathcal{O}_{er} \\$$





M. Swoboda et al., PRL 104, 103003 (2010)

# **RABBIT** with a resonant autoionizing state



 $N_2 + \Upsilon_{Hn} \pm \Upsilon_{IR} \rightarrow N_2^+(X) + e^-$ 

J. Caillat et al., PRL 106, 093002 (2011)

Resonant contribution still largely dominant

# **XUV** absorption with synchrotron radiation



R P Madden, K Codling, Ast. Phys. J. **141**, 364 (1965)

 $\operatorname{He} + \gamma \to \operatorname{He}^+(1s) + e^-$ 

# **RABITT** with an intermediate autoionizing state

He +  $\gamma_{\omega} \longrightarrow$  He<sup>+</sup>(1s) + e<sup>-</sup> He<sup>+</sup>(1s) + e<sup>-</sup> +  $\gamma \longrightarrow$  He<sup>+</sup>(1s) + e<sup>-</sup> He +  $\gamma_{\omega} \longrightarrow$  He<sup>\*\*</sup> He<sup>\*\*</sup> He<sup>\*\*</sup> +  $\gamma \longrightarrow$  He<sup>+</sup>(1s) + e<sup>-</sup>



I. What should we expect when both the bound and the continuum part of an intermediate resonant state contribute to the transition?

2. How does a finite duration of the pulse, comparable with the resonance lifetime, affects the measurement?

3. How is the electron dynamics to be interpreted?

 $I_{\rm SB}(E_f;\omega_{\rm IR},\tau) = \left|\mathcal{A}^+(\omega_{\rm IR},\tau)\right|^2 + \left|\mathcal{A}^-(\omega_{\rm IR},\tau)\right|^2 + 2\,\Re e\left[\mathcal{A}^{+*}(\omega_{\rm IR},\tau)\mathcal{A}^-(\omega_{\rm IR},\tau)\right]$ 



 $I_{\rm SB}(E_f;\omega_{\rm IR},\tau) = \left|\mathcal{A}^+(\omega_{\rm IR},\tau)\right|^2 + \left|\mathcal{A}^-(\omega_{\rm IR},\tau)\right|^2 + 2\,\Re e\left[\mathcal{A}^{+*}(\omega_{\rm IR},\tau)\mathcal{A}^-(\omega_{\rm IR},\tau)\right]$ 



There certainly is a shift. Yet, something is not quite right ...

 $I_{\rm SB}(E_f;\omega_{\rm IR},\tau) = \left|\mathcal{A}^+(\omega_{\rm IR},\tau)\right|^2 + \left|\mathcal{A}^-(\omega_{\rm IR},\tau)\right|^2 + 2\,\Re e\left[\mathcal{A}^{+*}(\omega_{\rm IR},\tau)\mathcal{A}^-(\omega_{\rm IR},\tau)\right]$ 



The "shift" increases with the time delay The RABBIT frequency itself is altered

## More than one way to skin a cat



# Fano recap



# Time-resolved resonant two-photon model

$$\mathcal{A}_{f\leftarrow i}^{(2)} = -\mathrm{i} \int_{-\infty}^{\infty} d\omega \tilde{A}(\omega_{fi} - \omega) \tilde{A}(\omega) \mathcal{M}_{fi}(\omega) \qquad \tilde{A}(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \mathcal{A}(t) e^{i\omega t} dt$$

 $\mathcal{M}_{fi}(\omega) = \alpha^2 \langle \psi_{\beta E_f} | P_z G_0^+(\omega_i + \omega) P_z | \psi_i \rangle \qquad G_0^+(E) \equiv (E - H + i0^+)^{-1}$ 

$$\mathcal{M}_{fi}(\omega) = \alpha^2 \oint d\varepsilon \frac{\langle \psi_{E_f \ell m} | P_z | \psi_{\varepsilon 10} \rangle \langle \psi_{\varepsilon 10} | P_z | \psi_i \rangle}{\omega_i + \omega - \varepsilon + \mathrm{i}0^+}$$

$$\begin{split} |\psi_E\rangle &= |E\rangle + |a\rangle \frac{V_{aE}}{E - \tilde{E}_a} + \int d\varepsilon |\varepsilon\rangle \frac{V_{\varepsilon a}}{E - \varepsilon + i0^+} \frac{V_{aE}}{E - \tilde{E}_a} \\ \mathbf{H} \qquad \mathbf{D} \qquad \mathbf{M} \end{split}$$

$$\mathcal{M}_{f\leftarrow i}(\omega) = \sum_{\alpha,\beta,\gamma\in\{D,M,H\}} \mathcal{M}_{\gamma\leftarrow\beta\leftarrow-\alpha\leftarrow i}(\omega) + \sum_{\gamma\in\{D,M,H\}} \mathcal{M}_{\gamma\leftarrow B\leftarrow i}(\omega)$$

Á. Jiménez Galán, L. Argenti, F. Martín, PRL 113, 263001 (2014)

# Time-resolved resonant two-photon model

$$\begin{split} \sum_{\alpha} \mathcal{M}_{H \leftarrow M \leftarrow \alpha \leftarrow g}(\omega) &= \int d\varepsilon \frac{\langle E_f | \mathcal{O} | \psi_{\varepsilon,M} \rangle \mathcal{O}_{\varepsilon g}}{E_g + \omega - \varepsilon + i0^+} \frac{\epsilon + q}{\epsilon - i} & \text{(f)} \\ \langle E_f | \mathcal{O} | \psi_{\varepsilon,M} \rangle &= \int d\varepsilon' \langle E_f | \mathcal{O} | \varepsilon' \rangle \frac{V_{\varepsilon'a}}{\varepsilon - \varepsilon' + i0^+} \frac{V_{a\varepsilon}}{\varepsilon - \tilde{E}_a} & \text{(f)} \\ \langle E_f | \mathcal{O} | \varepsilon' \rangle &\approx \bar{\mathcal{O}}_{fE} \delta(\varepsilon' - E_f) & \text{(f)} \\ \frac{1}{a - b + i0^+} \frac{1}{b - c + i0^+} &= \frac{1}{a - c + i0^+} \left[ \frac{1}{a - b + i0^+} + \frac{1}{b - c + i0^+} \right] \\ P \int d\varepsilon \frac{\mathcal{O}_{\varepsilon g}}{\varepsilon - E_f} - P \int d\varepsilon \frac{\mathcal{O}_{\varepsilon g}}{\varepsilon - (E_g + \omega)} - i\pi \mathcal{O}_{E_f g} - i\pi \mathcal{O}_{E_g + \omega, g} \end{split}$$

Á. Jiménez Galán, L. Argenti, F. Martín, PRL 113, 263001 (2014)
C. Marante, L. Argenti and F. Martín, Phys. Rev. A 90, 012506 (2014)

# Time-resolved resonant two-photon model

$$\mathcal{A}_{f\leftarrow i}^{(2)} = -i \int_{-\infty}^{\infty} d\omega \tilde{A}(\omega_{fi} - \omega) \tilde{A}(\omega) \mathcal{M}_{fi}(\omega)$$
$$A(t) = A_0 e^{-\frac{1}{2}\sigma^2(t - t_0)^2} \cos[\omega_0(t - t_0) + \phi]$$
$$(t, \tau) = A_{APT}(t) + A_{IR}(t; \tau), \qquad A_{APT}(t) = \sum_n A_{H_{2n+1}}(t)$$

$$\mathcal{A}_{f_H \leftarrow i}^{(2),\pm} = \mathcal{F}(\tau) \left[ w(z_f^{\pm}) + \left(\beta_{Ea} - \epsilon_{fa}^{-1}\right) \left(q_a' - i\right) w(z_a^{\pm}) \right]$$

$$z_{a}^{\pm} = \frac{\sigma_{t}}{\sqrt{2}} (\tilde{\omega}^{\pm} - \omega_{\tilde{a}i}) \qquad \tilde{\omega}^{\pm} = \omega_{\mathrm{H}_{2n\pm1}} + \frac{\delta_{f}}{\sigma_{t}^{2}\sigma_{\mathrm{IR}}^{2}} - i\frac{\tau}{\sigma_{t}^{2}} \qquad \delta_{f} = E_{f} - E_{\mathrm{SB}_{2n}}$$
$$z_{f}^{\pm} = \frac{\sigma_{t}}{\sqrt{2}} (\tilde{\omega}^{\pm} - \omega_{fi}) \qquad w(z) = e^{-z^{2}} \left( 1 + \frac{2i}{\sqrt{\pi}} \int_{0}^{z} e^{t^{2}} dt \right) \qquad \begin{array}{c} \mathsf{Faddeeva} \\ \mathsf{function} \end{array}$$

 $\beta_{Ea} = \sqrt{\frac{\pi\Gamma}{2}} \frac{\mathcal{O}_{Ea}}{\bar{\mathcal{O}}_E}$ 

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Á. Jiménez Galán, L. Argenti, F. Martín, PRL 113, 263001 (2014)

# Non-resonant RABBIT red-shift

$$\langle E_s | \mathcal{O}G_0^+(E_g + \omega_{2n\pm 1})\mathcal{O}|g\rangle \simeq \pm \frac{1}{\omega_{\mathrm{IR}}} \left( \int_{E_s - \delta}^{E_s + \delta} d\epsilon \langle E_s | \mathcal{O}|\epsilon_p \rangle \right) \langle E_p | \mathcal{O}|g\rangle$$



#### Approx. the same for all sidebands

Á. Jiménez Galán, F. Martín, L. Argenti, *manuscript in preparation* (2015)
C. Marante, L. Argenti and F. Martín, Phys. Rev. A 90, 012506 (2014)

 $I_{\rm SB}(E_f;\omega_{\rm IR},\tau) = \left|\mathcal{A}^+(\omega_{\rm IR},\tau)\right|^2 + \left|\mathcal{A}^-(\omega_{\rm IR},\tau)\right|^2 + 2\,\Re e\left[\mathcal{A}^{+*}(\omega_{\rm IR},\tau)\mathcal{A}^-(\omega_{\rm IR},\tau)\right]$ 



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Á. Jiménez Galán, L. Argenti, F. Martín, PRL 113, 263001 (2014)

$$\langle E|\mathcal{O}G_0^+(E_g + \omega_{\rm XUV})\mathcal{O}|g\rangle \simeq \frac{\langle E|\mathcal{O}|a\rangle\langle a|\mathcal{O}|g\rangle}{\omega_{\rm XUV} - E_{ag} + i\Gamma_a/2} + \mathcal{M}_{\rm dir}(E)$$

$$\langle E|\mathcal{O}G_0^+(E_g + \omega_{\rm XUV})\mathcal{O}|g\rangle \simeq \frac{\langle E|\mathcal{O}|a\rangle\langle a|\mathcal{O}|g\rangle}{\omega_{\rm XUV} - E_{ag} + i\Gamma_a/2}$$
  $\frac{1}{x-z}$ 

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  $\frac{1}{x-z}$ 



$$\left|\frac{1}{x-z} - \frac{i}{2z_I}\right|^2 = \frac{1}{(2z_I)^2}$$

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  $\frac{1}{x-z}$ 



$$\varphi(x) = \arg\left(\frac{1}{x-z}\right) = \arctan\left(\frac{x-z_R}{|z_I|}\right) - \frac{\pi}{2}$$

$$\langle E|\mathcal{O}G_0^+(E_g + \omega_{\rm XUV})\mathcal{O}|g\rangle \simeq \frac{\langle E|\mathcal{O}|a\rangle\langle a|\mathcal{O}|g\rangle}{\omega_{\rm XUV} - E_{ag} + i\Gamma_a/2} + \mathcal{M}_{\rm dir}(E)$$



$$\delta\varphi(\omega_{\mathrm{IR}},\tau) = -\arg\left\{w^*(z_f^-)\left[B\,w(z_a^+) - w(z_f^+)\right]\right\}$$

$$B = [\beta_{Ea} - 2^{-1}\Gamma_a / (E_f - E_a)](i - q'_a)$$

# **Apparent phase modulation**





Us from Madrid, plus other people, work in preparation (2015)



Álvaro, Fernando and I, work in preparation (2015)



1. Close to resonance, exponentially decaying SB appears

Álvaro, Fernando and I, work in preparation (2015)



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Álvaro, Fernando and I, work in preparation (2015)



# Separated pump-probe

1. sideband beating persists beyond PP overlap



# Separated pump-probe

1. sideband beating persists beyond PP

2. change of phase and frequency wrt to "normal" RABBIT

3. complex q parameter for final resonance

4. Inversion of Fano

10:35 Á. Jiménez Galán, L. Argenti, F. Martín, PRL 113, 263001 (2014)

# **Resonant RABITT ionization of Argon**



M. Kotur et al., arXiv:1505.02024 [physics.atom-ph] (2015)

10:35









$$\begin{split} |\varphi_D\rangle &= |\varphi_{[3p^{-1}\otimes\varepsilon_s]_P}\rangle V_d/V - |\varphi_{[3p^{-1}\otimes\varepsilon_d]_P}\rangle V_s/V \\ |\varphi_R\rangle &= |\varphi_{[3p^{-1}\otimes\varepsilon_s]_P}\rangle V_s/V + |\varphi_{[3p^{-1}\otimes\varepsilon_d]_P}\rangle V_d/V \end{split}$$

Unitary Transf.



$$\begin{aligned} |\varphi_D\rangle &= |\varphi_{[3p^{-1}\otimes\varepsilon_s]_P}\rangle V_d/V - |\varphi_{[3p^{-1}\otimes\varepsilon_d]_P}\rangle V_s/V & Unitary \\ |\varphi_R\rangle &= |\varphi_{[3p^{-1}\otimes\varepsilon_s]_P}\rangle V_s/V + |\varphi_{[3p^{-1}\otimes\varepsilon_d]_P}\rangle V_d/V & Transf. \end{aligned}$$

 $|\varphi_R\rangle \qquad |\varphi_D\rangle$   $V_R \qquad V_D \quad 3s^{-1}4p \qquad V_R = \langle \varphi_R | H | 3s^{-1}4p \rangle = V$   $V_D = \langle \varphi_D | H | 3s^{-1}4p \rangle = 0$ 

# Decoupling of intermediate multichannel-resonant states



# Decoupling of intermediate multichannel-resonant states



# Decoupling of intermediate multichannel-resonant states



 $\mathcal{A}_{2n,\,\beta\leftarrow R\leftarrow g}^{\pm} = e^{\mp i\omega_{\mathrm{IR}}\tau}\mathcal{F}_R(\tau) \left[ w(z_f^{\pm}) + (\beta_{Ea} - \epsilon_{fa}^{-1})(q_R - i)w(z_a^{\pm}) \right]$ 

 $I_{2n}(\tau) = I_{2n, [3p^{-1} \otimes E_p]_S \leftarrow g}(\tau) + I_{2n, [3p^{-1} \otimes E_p]_D \leftarrow g}(\tau) + I_{2n, [3p^{-1} \otimes E_f]_D \leftarrow g}(\tau)$ 





M. Kotur et al., arXiv:1505.02024 [physics.atom-ph] (2015)





M. Kotur et al., arXiv:1505.02024 [physics.atom-ph] (2015)

 $I_{2n}(\tau) = I_{2n, [3p^{-1} \otimes E_p]_S \leftarrow g}(\tau) + I_{2n, [3p^{-1} \otimes E_p]_D \leftarrow g}(\tau) + I_{2n, [3p^{-1} \otimes E_f]_D \leftarrow g}(\tau)$ 





M. Kotur et al., arXiv:1505.02024 [physics.atom-ph] (2015)

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M. Kotur et al., arXiv: 1505.02024 [physics.atom-ph] (2015)



# **Predictions Accounting for Blue Shift**



# Conclusion

A two-photon finite-pulse resonant model has been both necessary and sufficient to explain the phenomenology predicted numerically or observed experimentally so far.

The interpretation of the dynamics in terms of wavepackets is probably still viable but it cannot rely solely on stationary descriptions.

# Credit

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<u>Álvaro Jiménez Galán</u> Fernando Martín Alfred Maquet Eva Lindroth Markus Dahlström Anne L'Huillier <u>Marija Kotur</u> Diego Guénot David Kroon Mathieu Gisselbrecht



# **XCHEM code**



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10:50

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