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Nonlinear resonant Auger spectroscopy in CO using an x-ray pump-control scheme

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Acknowledgements

Victor Kimberg
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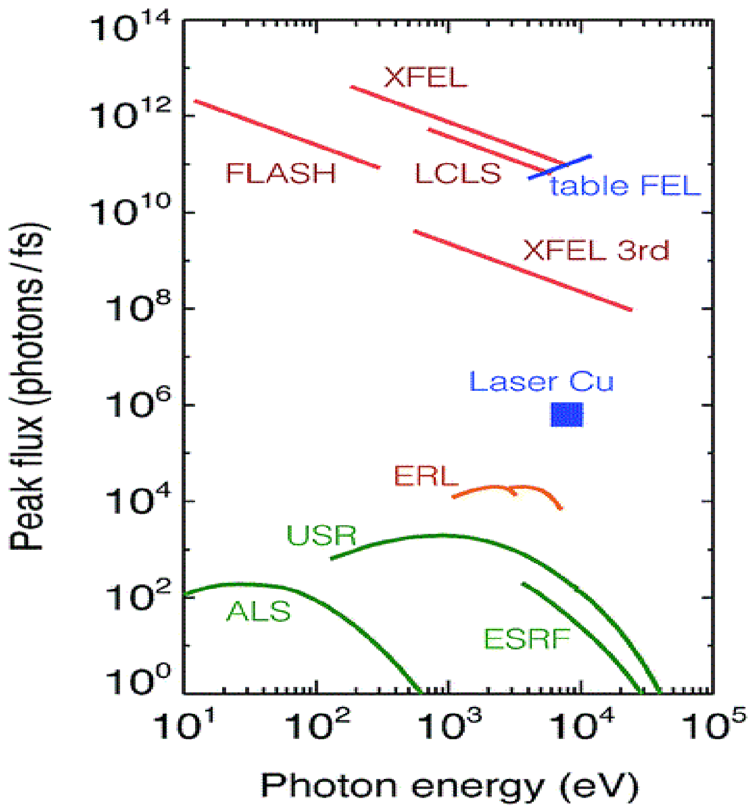
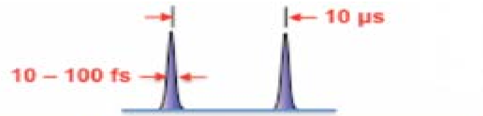
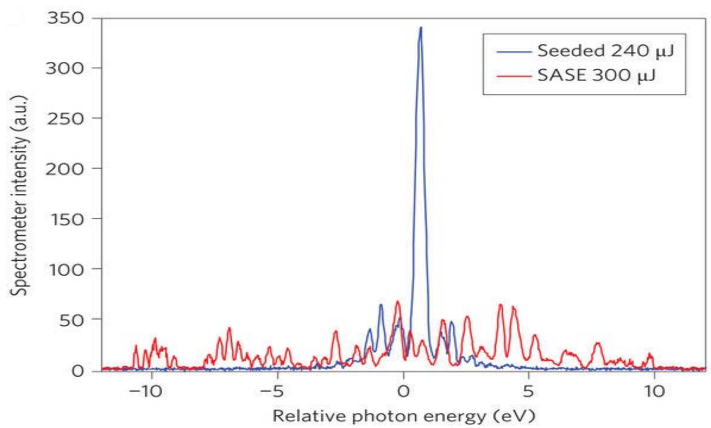
Motivations

Ultra intense & ultra short & multi color x-ray free electron lasers



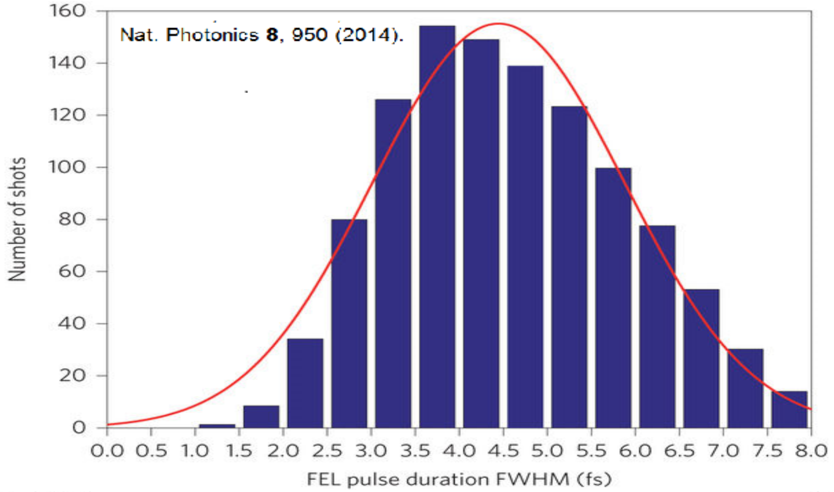


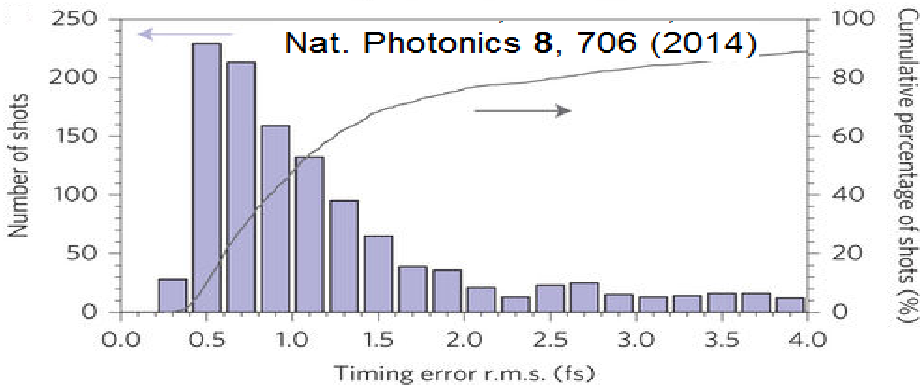
FEL	High resolution
Method	Laser-seeded Time/bandwidth limited
ph / pulse	$10^{10} – 10^{12}$
Pulse width	10 – 100 fs
Rate	0.1 – 1 MHz
Note	$10^{-3} – 10^{-4}$ bandwidth

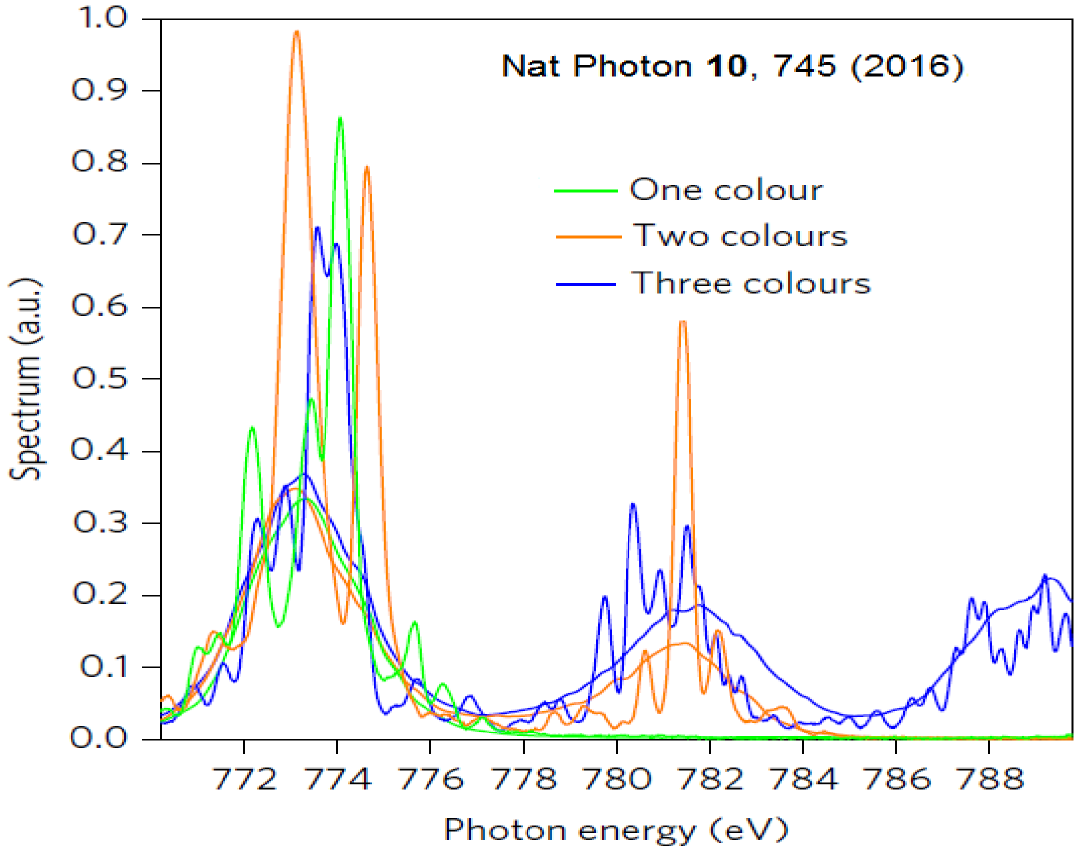


Motivations

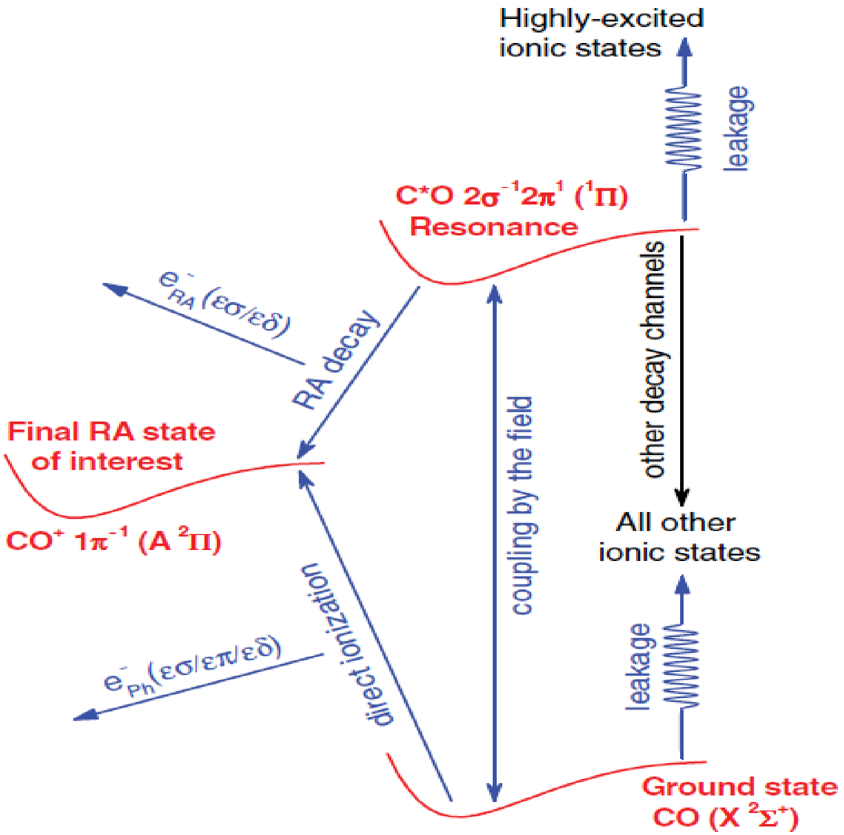
Ultra intense & ultra short & multi color x-ray free electron lasers

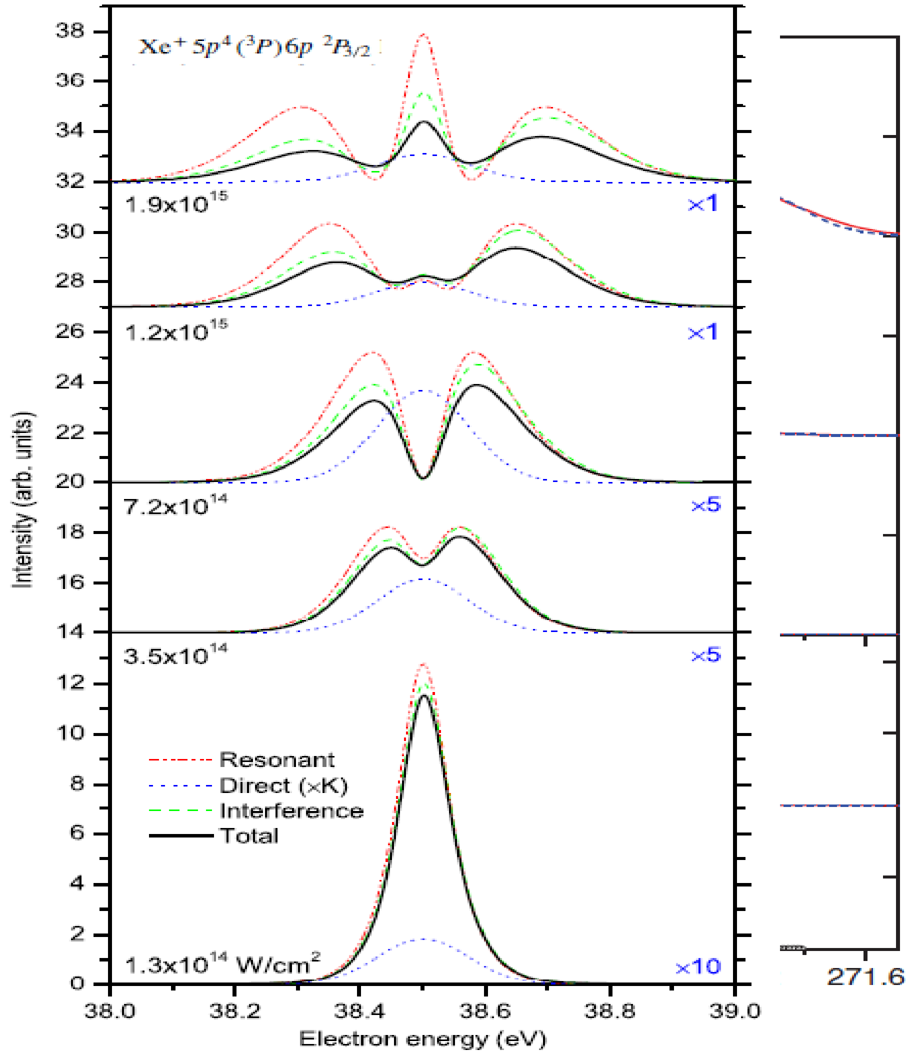






RAS in intense x-ray laser fields





Demekhin et al. Phys. Rev. A **84**, 033417 (2011)

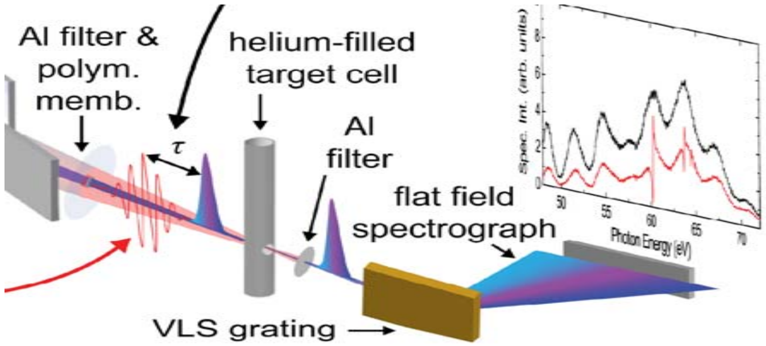
Müller & Demekhin, J. Phys. B **48**, 075602 (2015)

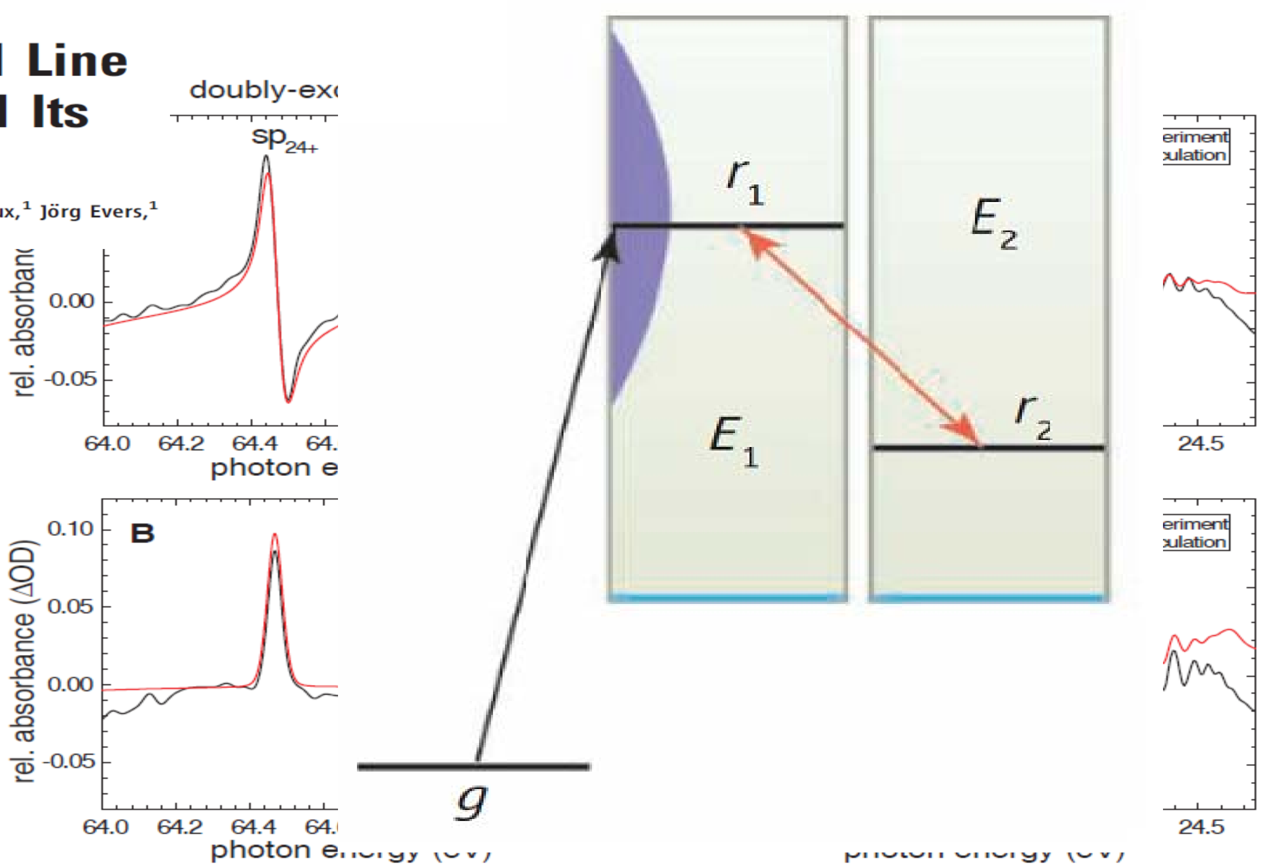
Coherent control or resonance

REPORTS

Lorentz Meets Fano in Spectral Line Shapes: A Universal Phase and Its Laser Control

Christian Ott,[1] Andreas Kaldun,[1] Philipp Raith,[1] Kristina Meyer,[1] Martin Laux,[1] Jörg Evers,[1] Christoph H. Keitel,[1] Chris H. Greene,[3] Thomas Pfeifer[1,2]*

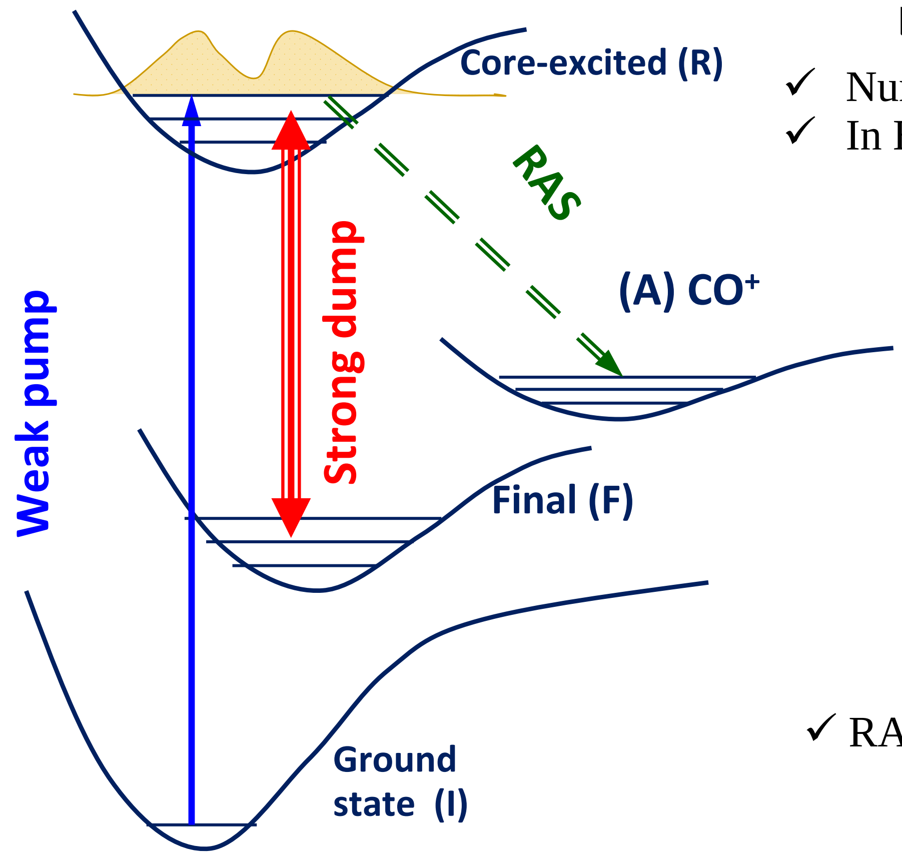




Science **340**, 716 (2013)

Science **340**, 694 (2013)

All x-ray pump-control scheme of RAS



- ❑Short (2 fs) delayed x-ray pulses
- ❑Population transfer and coherent control

✓ Numerical approach implemented in MCTDH

✓ In B-O approximation solving equations for vibrational wave function

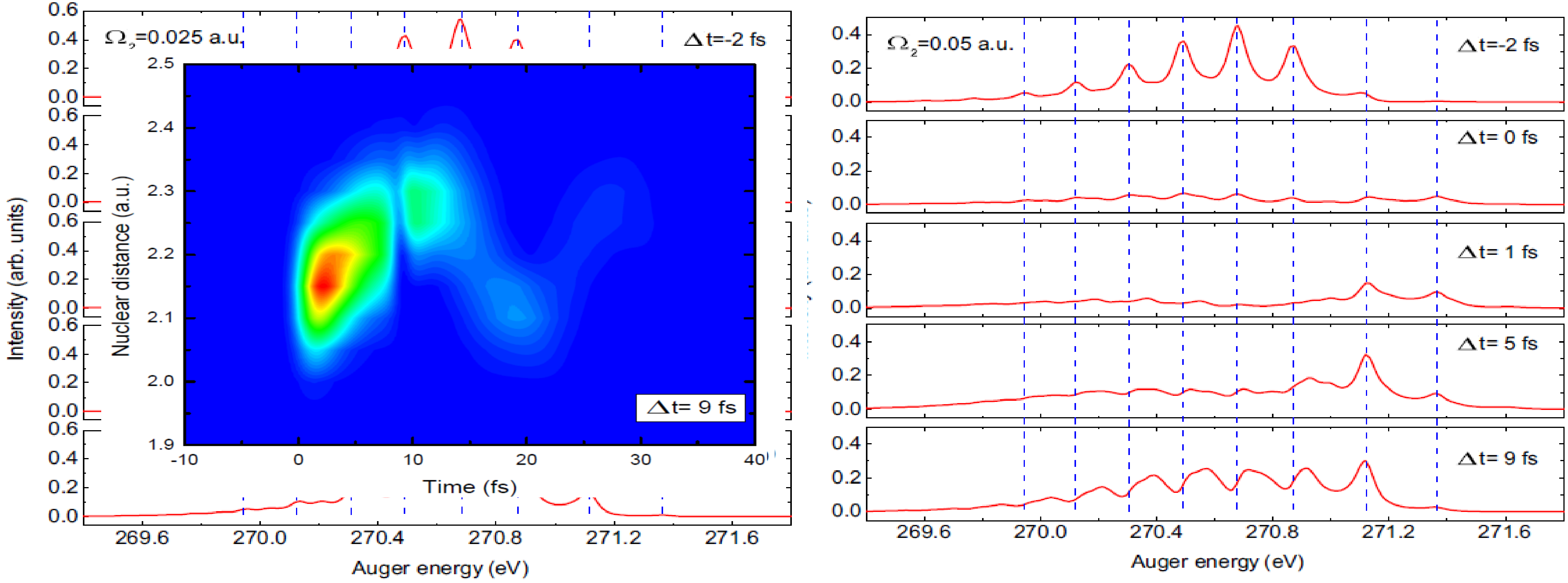
$$\begin{aligned}
\dot{\Psi}_I(t) &= \left[\widehat{\mathbf{T}}(R,\theta) + V_I(R) - \frac{i}{2}\Gamma_{\mathrm{ph}}(t)\right]\Psi_I(t) \\
&\quad + D^{\dagger}_{1x}(t)\sin\theta\,\Psi_R(t), \\
\dot{\Psi}_R(t) &= D_{1x}(t)\sin\theta\,\Psi_I(t) \\
&\quad + \left[\widehat{\mathbf{T}}(R,\theta) + V_R(R) - \frac{i}{2}\Gamma_{\mathrm{Aug}} - \omega_1\right]\Psi_R(t) \\
&\quad + D^{\dagger}_{2x}(t)\sin\theta\,\Psi_F(t), \\
\dot{\Psi}_F(t) &= D_{2x}(t)\sin\theta\,\Psi_R(t) \\
&\quad + [\widehat{\mathbf{T}}(R,\theta) + V_F(R) - (\omega_1 - \omega_2)]\Psi_F(t), \\
\dot{\Psi}_A(\varepsilon,t) &= V\Psi_R(t) + [\widehat{\mathbf{T}}(R,\theta) + V_A(R) + \varepsilon - \omega_1]\Psi_A(\varepsilon,t).
\end{aligned}$$

✓ RAS computed using the norm of wave packet in state (A)

$$\sigma_A(\varepsilon) = \lim_{t\to\infty}\langle\Psi_A(\varepsilon,t)|\Psi_A(\varepsilon,t)\rangle.$$

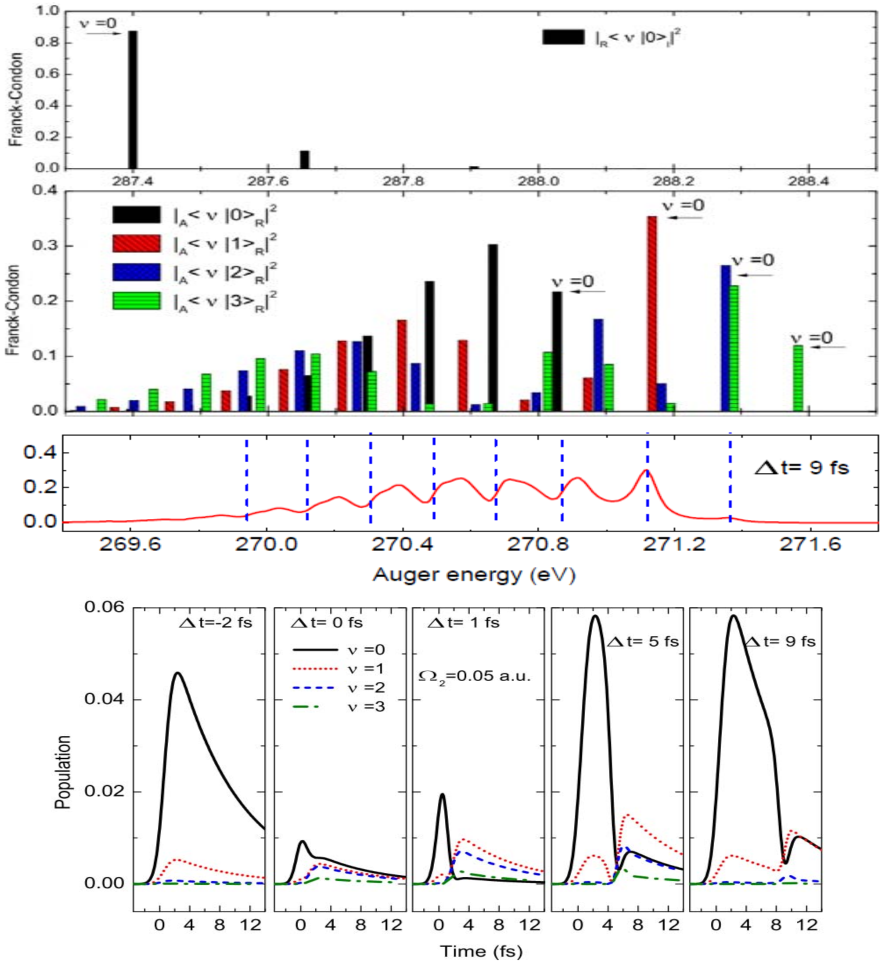
Zhang et al. Phys. Rev. A **94**, 063413 (2016)

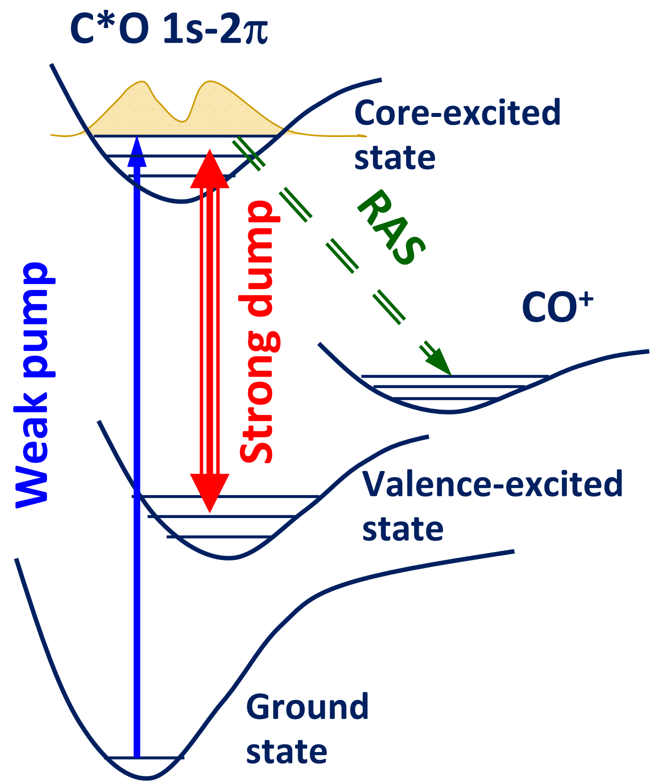
Time-delayed RAS for weak and strong control pulses



Zhang et al. Phys. Rev. A **94**, 063413 (2016)

Frank-Condon Analysis of RAS

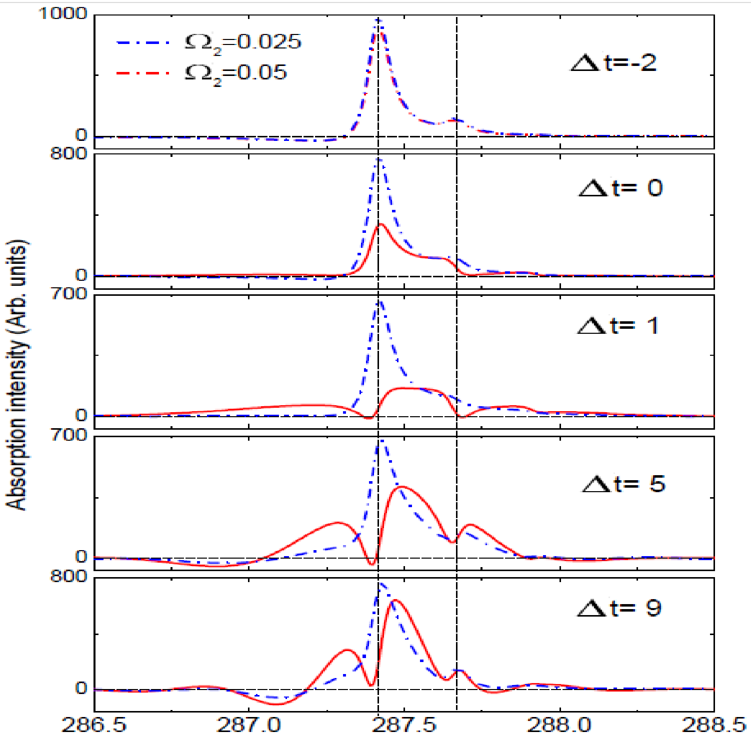




Zhang et al. Phys. Rev. A **94**, 063413 (2016)

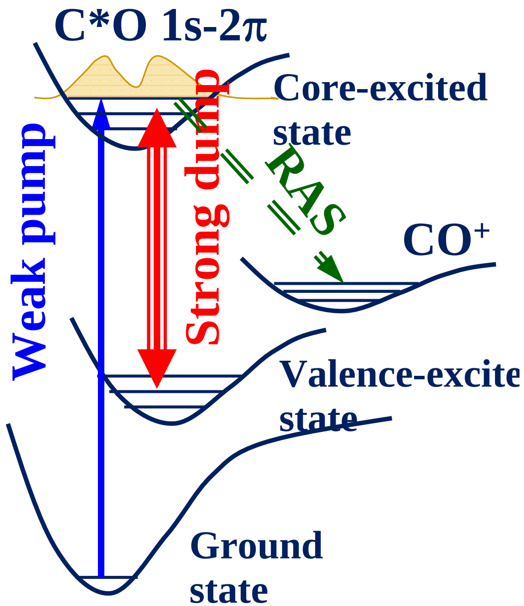
XAS and RAS are complementary techniques for monitoring the nonlinear dynamics

XAS

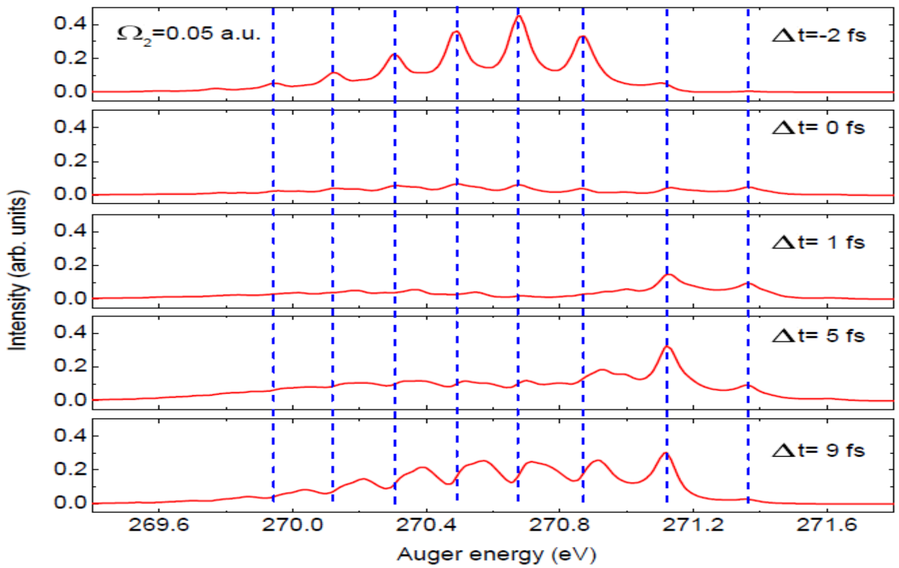


Pump energy (eV)

- RAS analysis is more clear for WP dynamics
- Independent channel
- Reach vibrational spectrum
- Easier experimental registration



RAS



$$S(\omega) = -2\mathrm{Im}[\mu(\omega)E^*(\omega)],$$

Zhang et al. Phys. Rev. A **94**, 063413 (2016)

Summary

A nonlinear femtosecond x-ray pump-probe scheme to study the vibrational dynamics of a core-excited molecular state is proposed.

Both RAS and transient absorption or emission spectra show strong sensitivity to the delay time between pulses.

With strong control pulse, the vibrational distribution of the upper state is shuffled by revolving the population via the resonantly coupled vibrationally excited state, which results in a coherent change of the wave packet in the core-excited state, with clear interference effects in the according RAS spectra.

Compared to transient absorption or emission spectroscopy of the weak pump pulses, the RAS spectra provide a clearer picture of the vibrational components involved.

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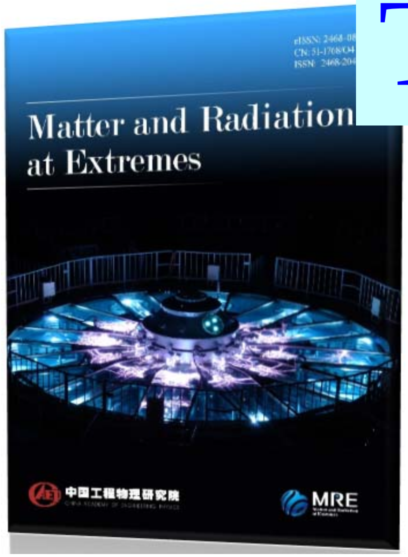

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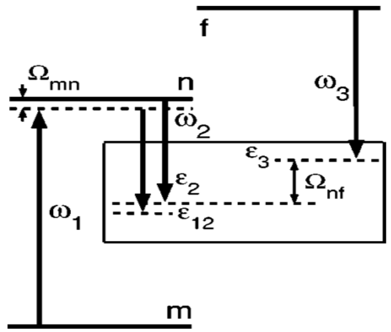
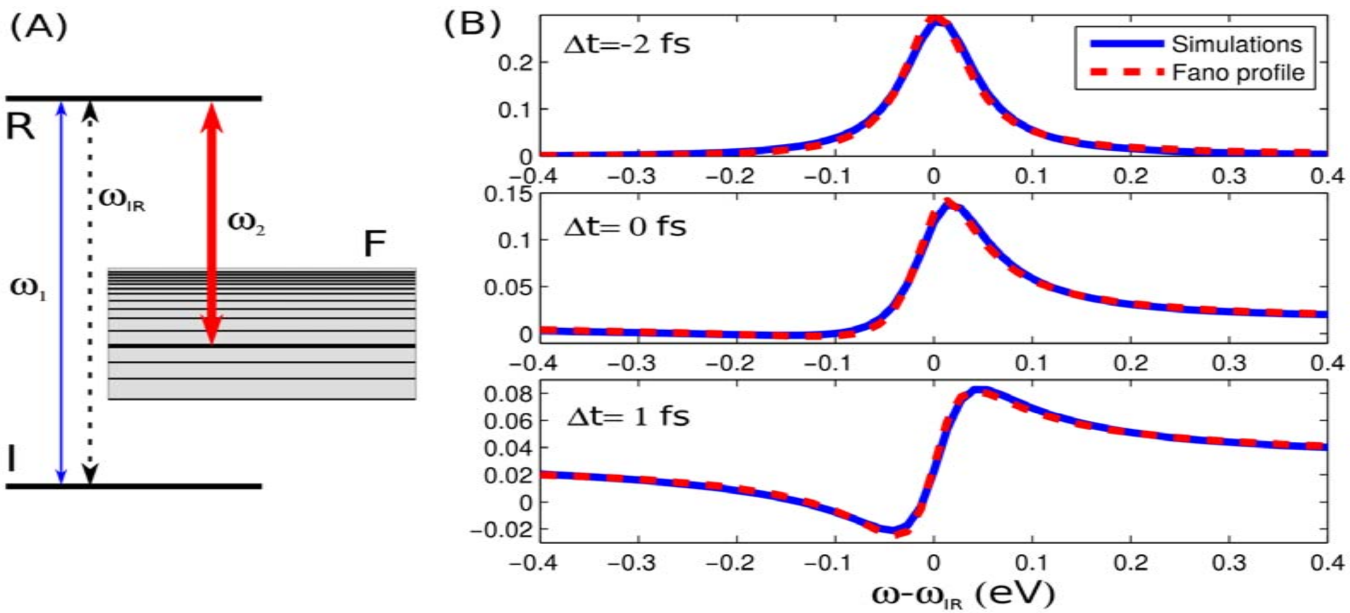
FANO LINE SHAPE

- Strong-field interaction with vibrational quasi-continuum
- Laser Induced Continuum Structure (LICS)[1]
- Pathways via LICS and quasi-continuum results in Fano-profile





$$\sigma(\varepsilon) = \sigma_0 \frac{(q+\varepsilon)^2}{1+\varepsilon^2}$$

$$\varepsilon = 2(\omega - \omega_{IR})/\Gamma_{Aug}$$

1) Popov, Kimberg, George, Phys Rev A, 68, 033407 (2003); 69, 043816 (2004)