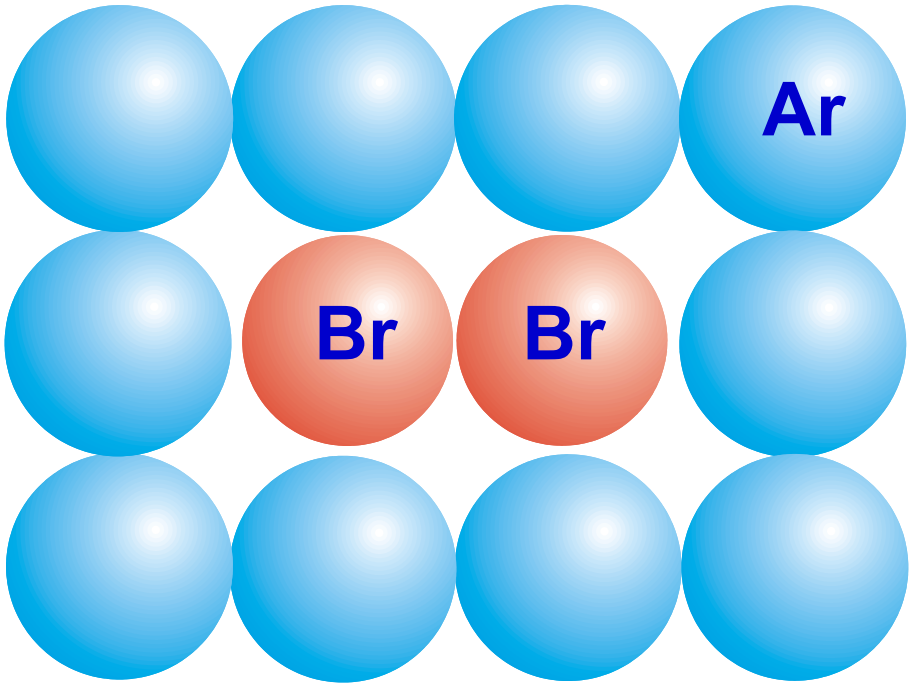
Perspectives for Time Resolved Structural Studies derived from Pump-Probe Spectroscopy



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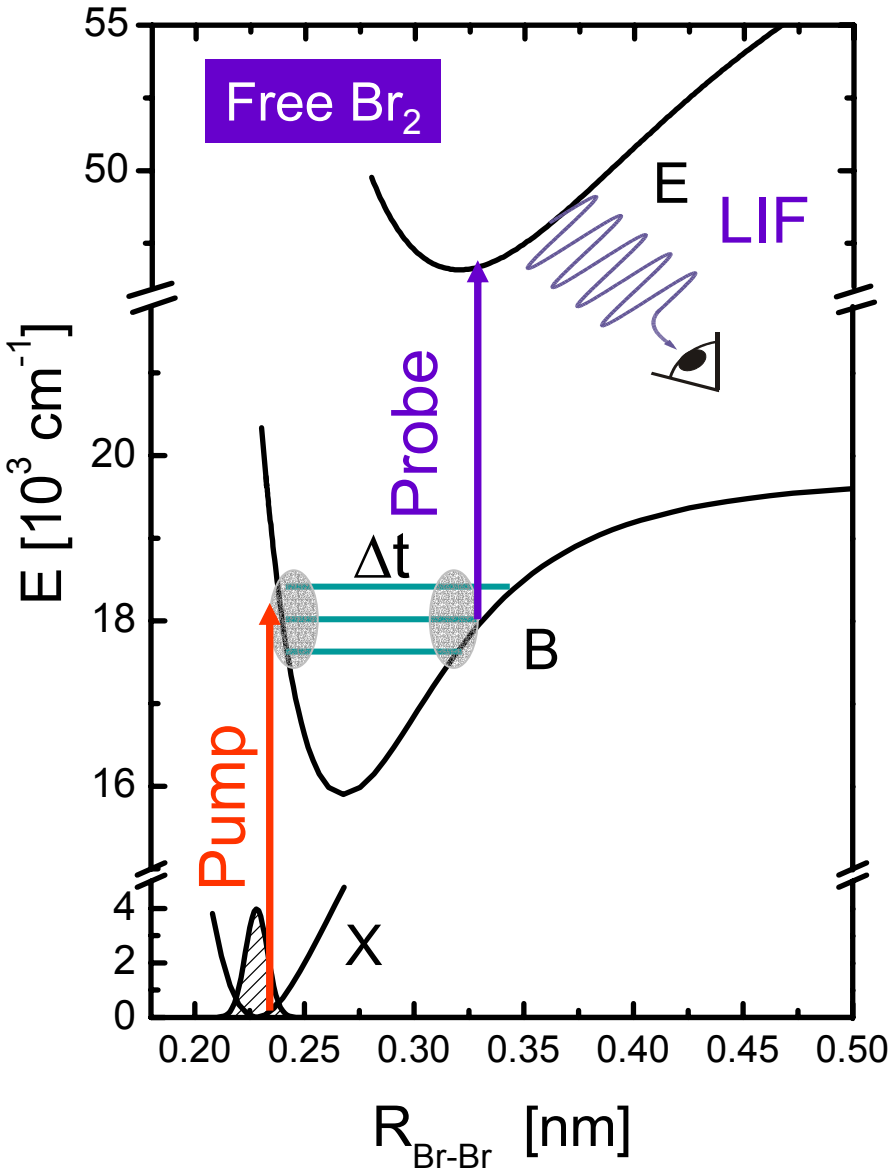
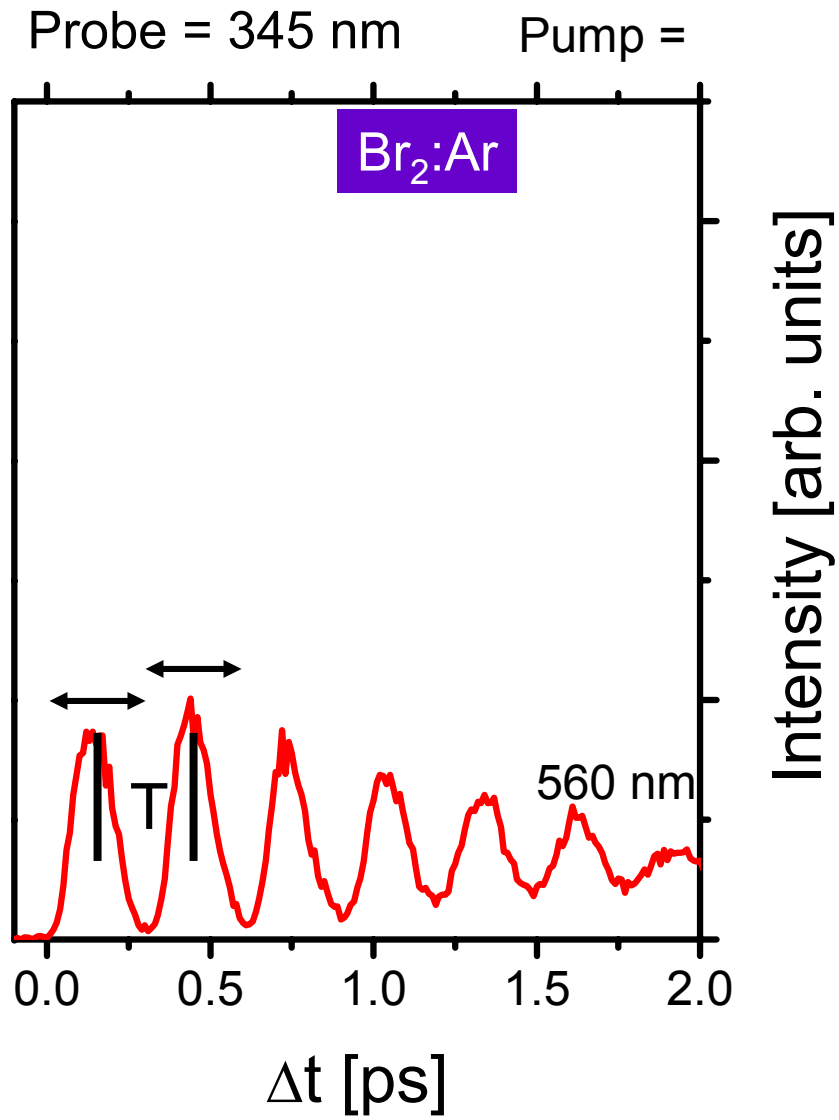
Outline

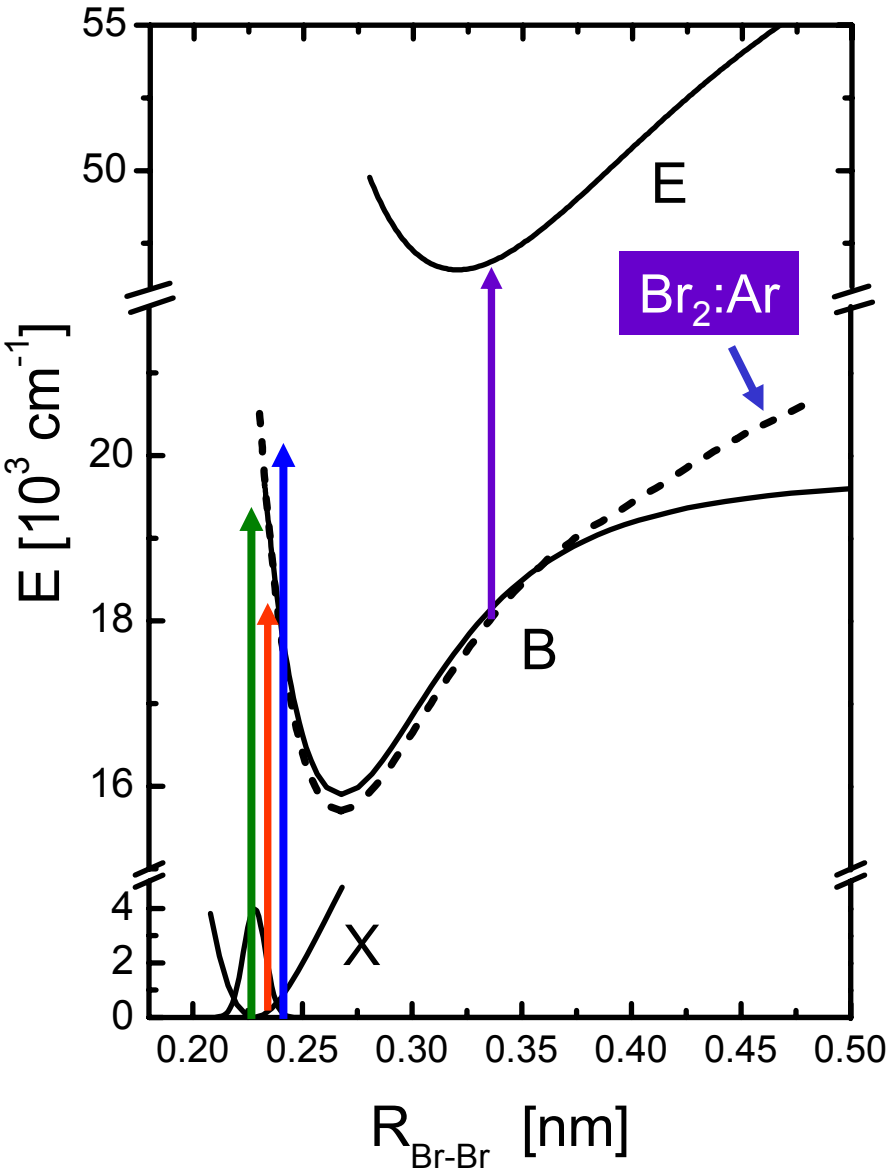
- Method: fs pump-probe spectroscopy
- Experimental RKR potential
- Revivals to learn about vibrational coherence
- Coherent control of revivals
- Coherent phonons

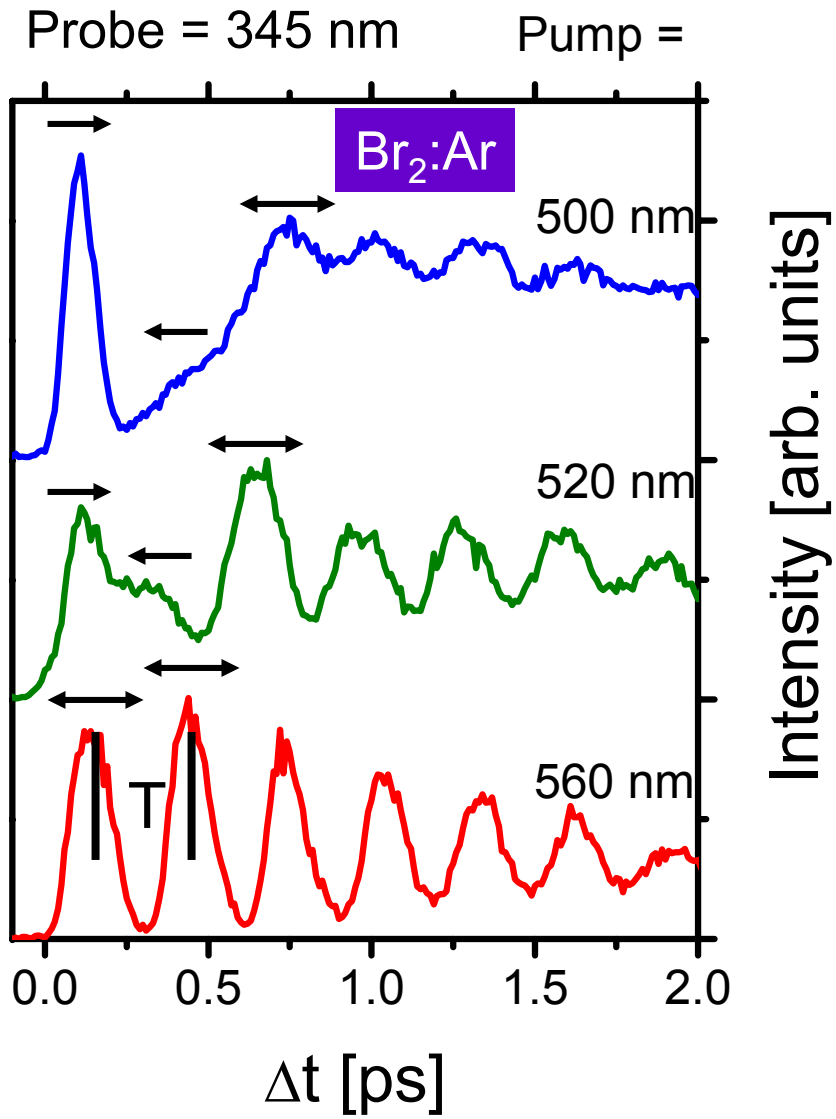
Method: Pump-probe spectroscopy



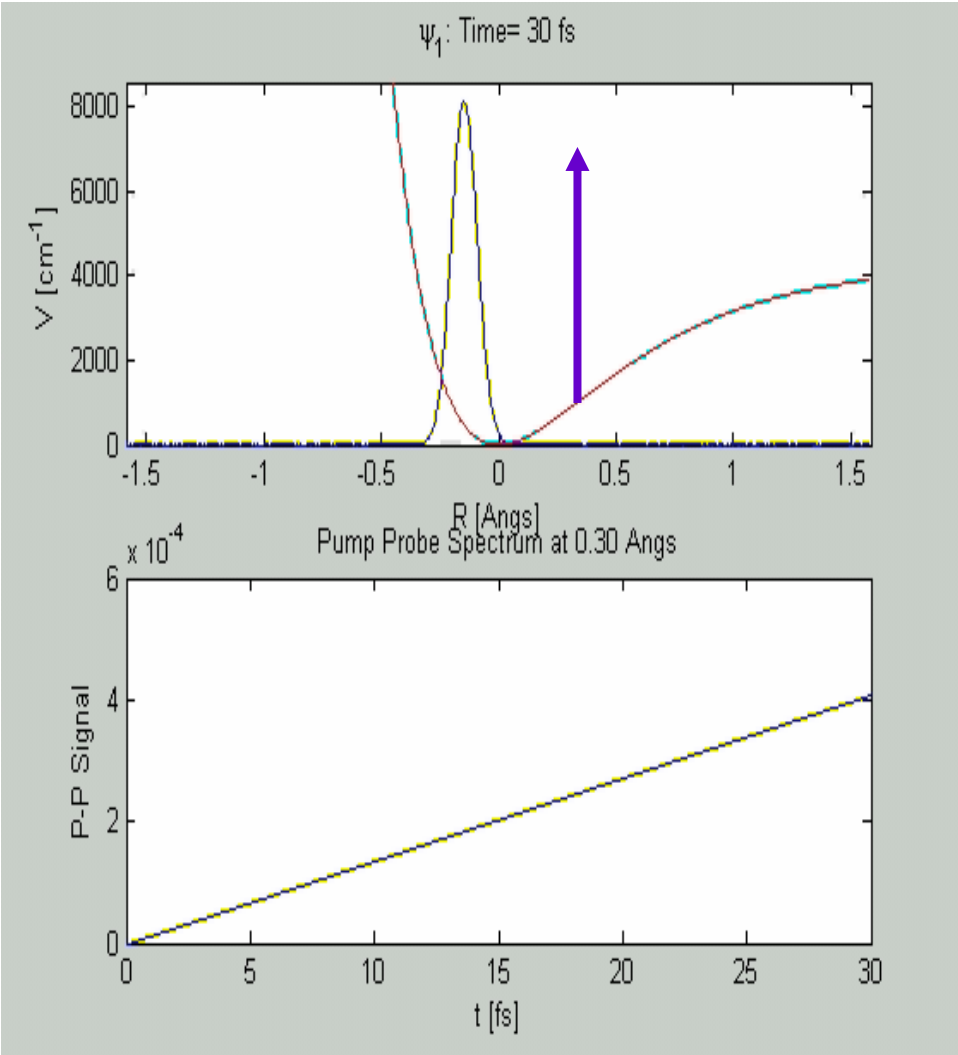


RKR Potential



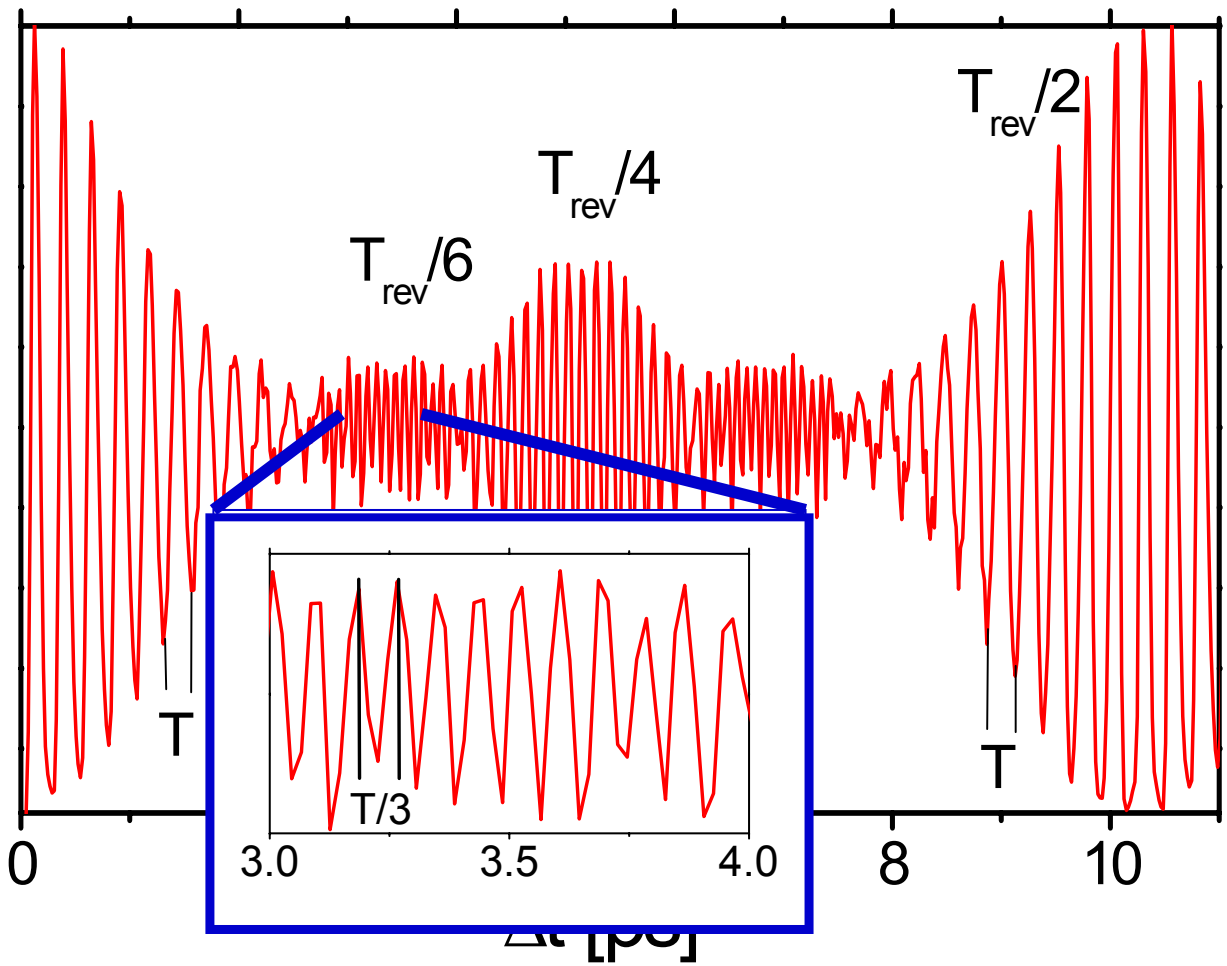


Dispersion of wave packets

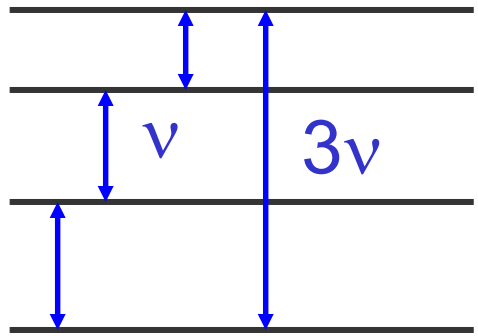


Anharmonic: WP broadens due to dispersion

Revivals of a vibrational wave packet



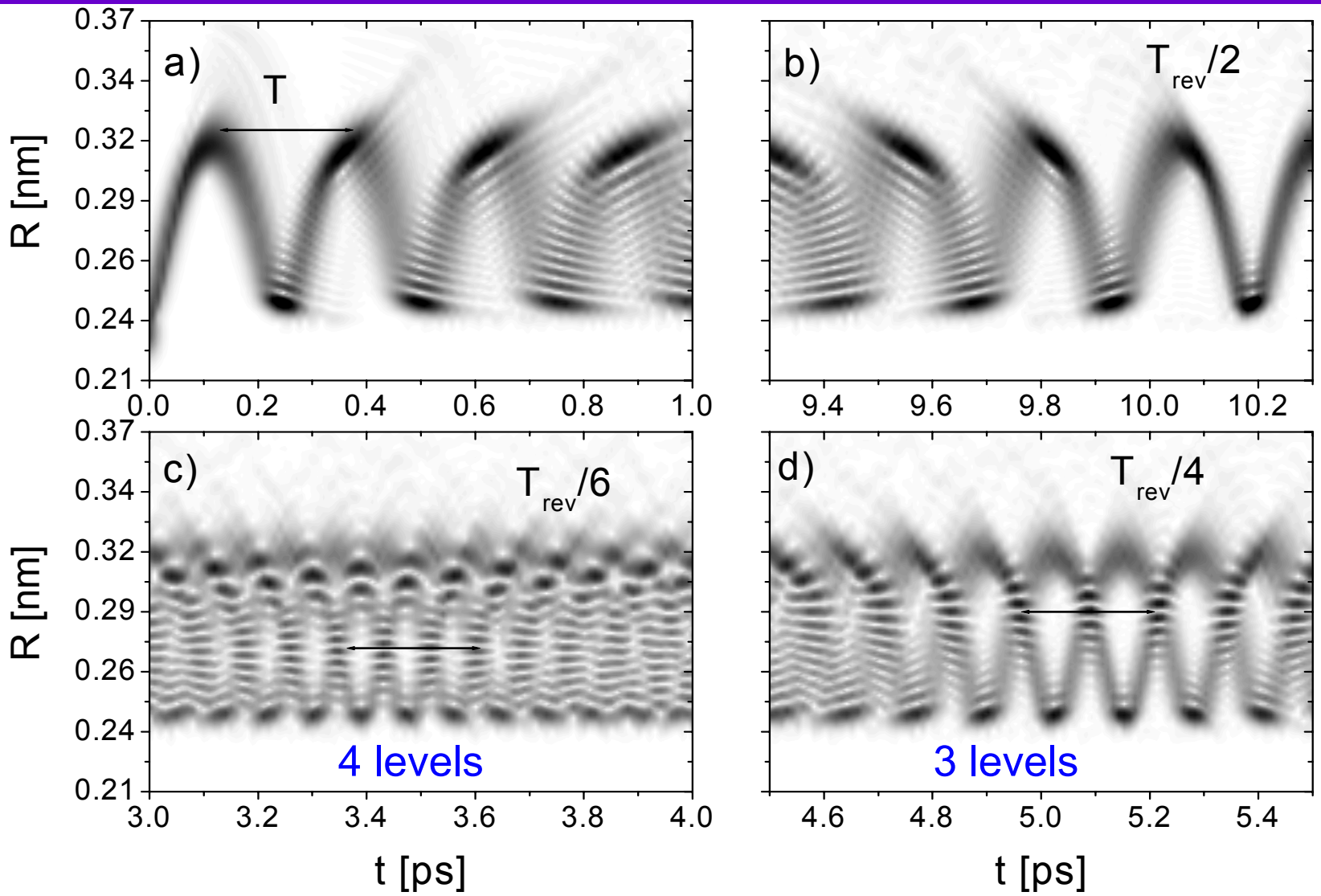
T_{rev} /6 at 3.5 ps
Frequency: 3ν



$T_{rev} = 2\pi/\omega_e x_e = 21.08$ ps for B of Br_2

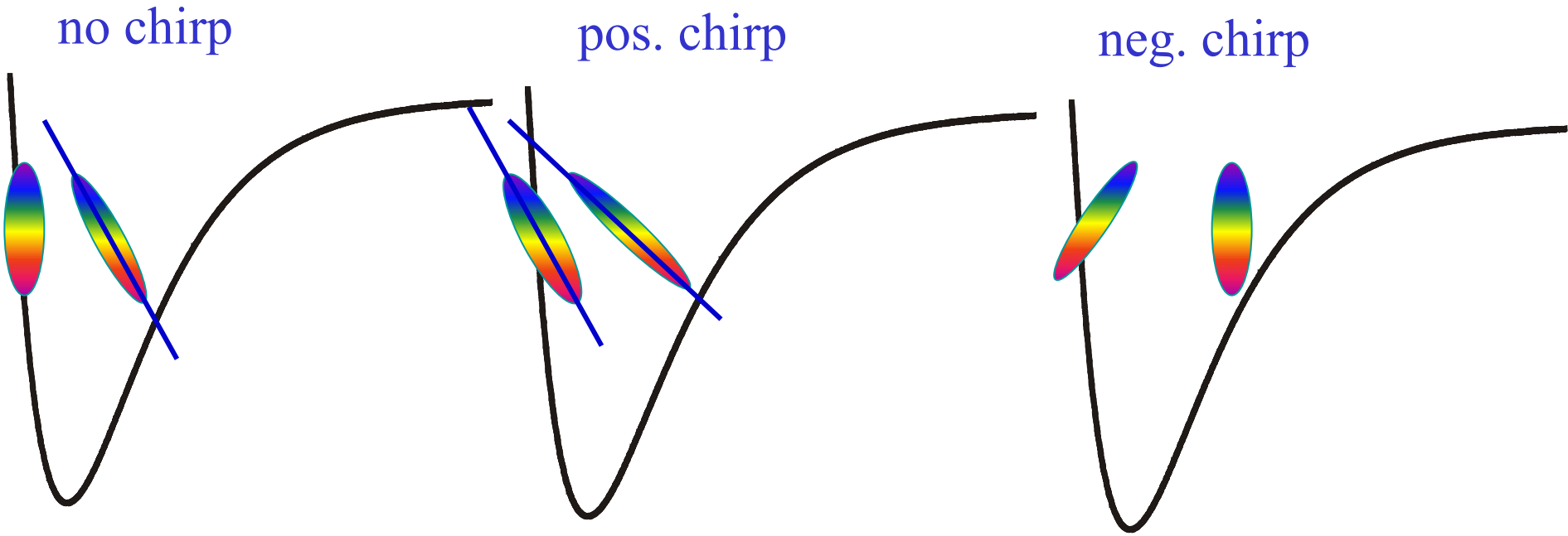
I. Sh. Averbukh *et al.*, Sov. Phys. Usp., **34**, 572 (1991)

Dispersion of wave packets



Vibrational coherence time: 3 ps

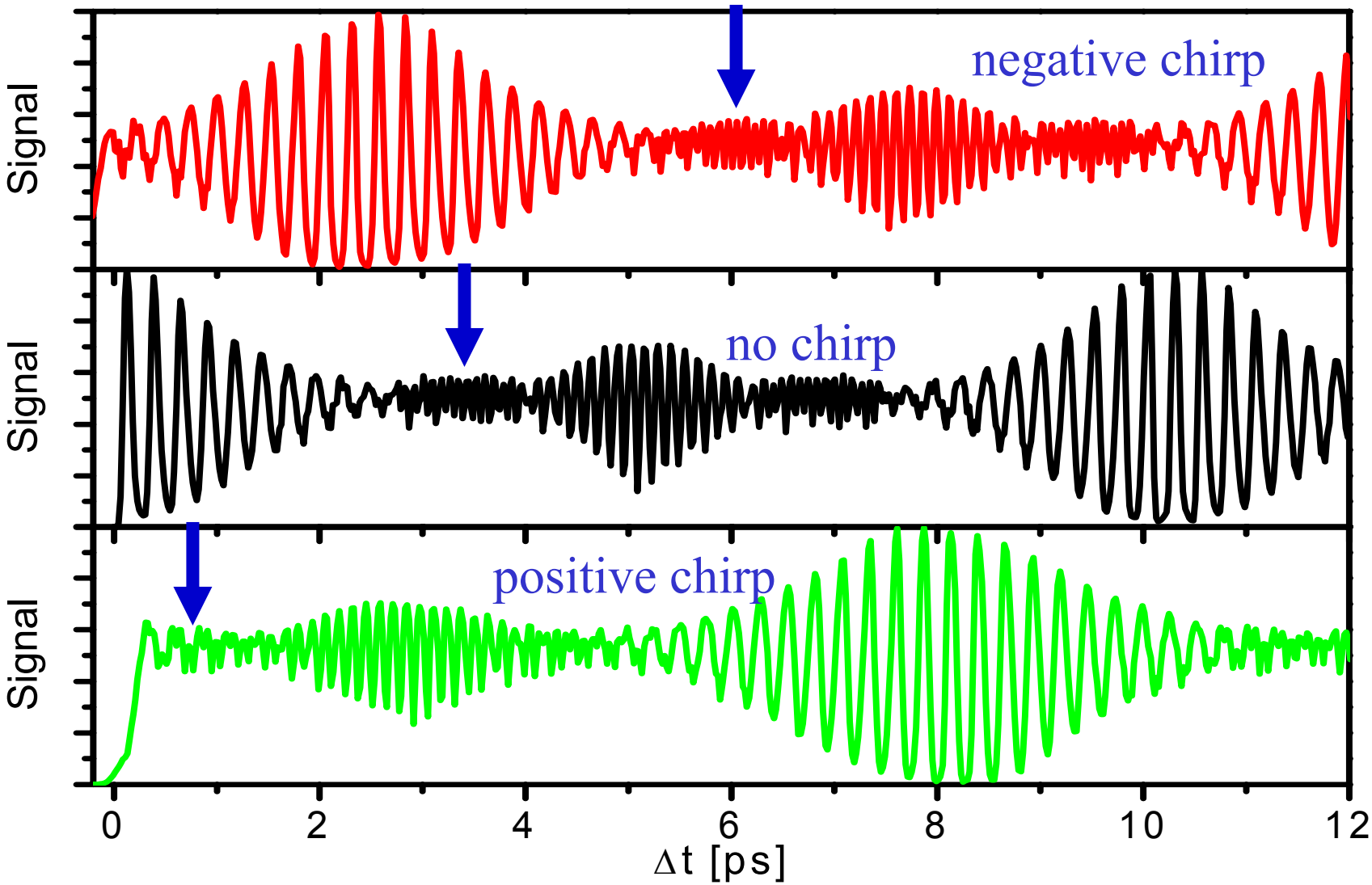
Coherent Control of Revival Structure



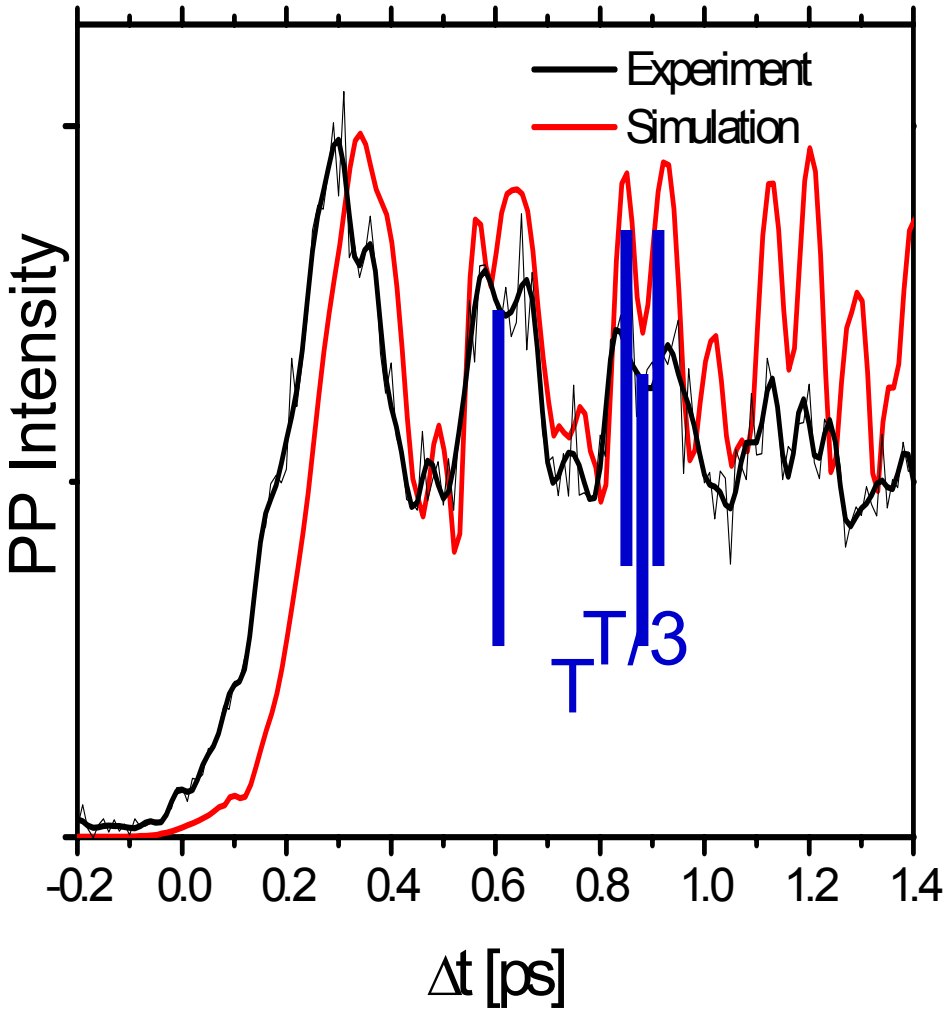
- chirp parameter determines appearance of revival

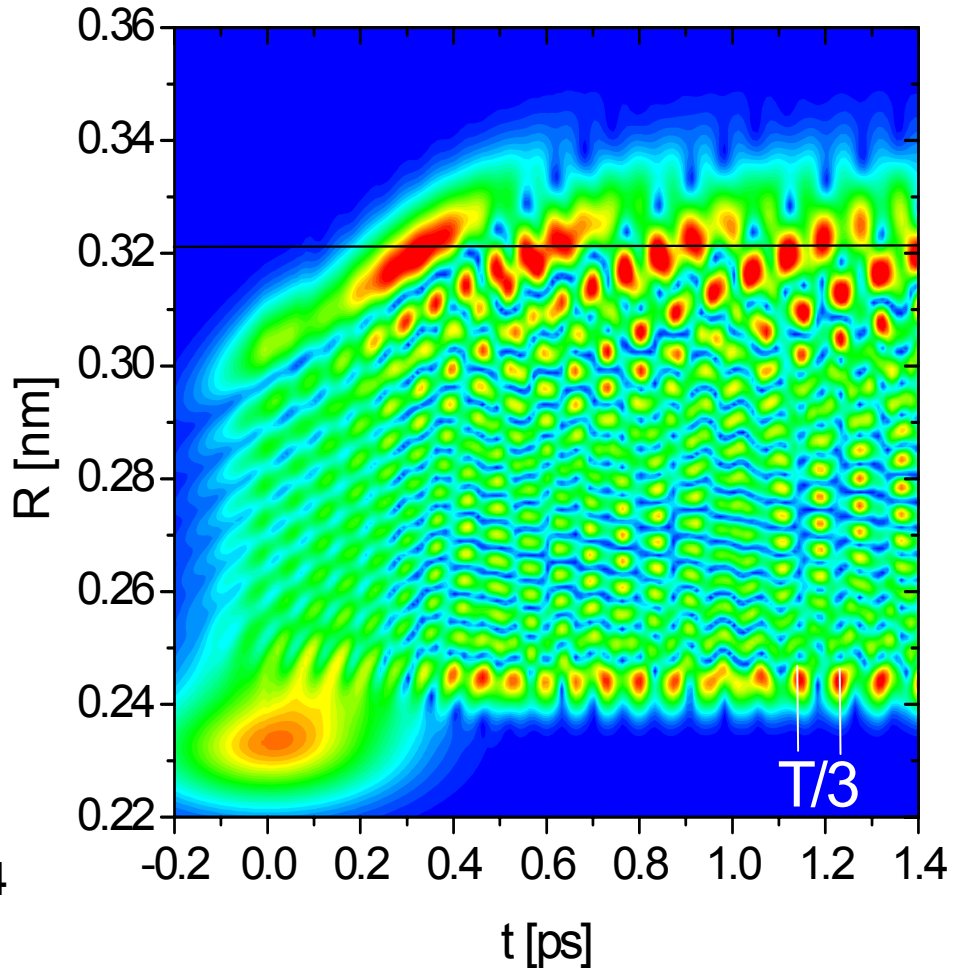
Gühr, Ibrahim, Schwentner, PCCP **6**, 5353-5361 (2004)

Control of Revival Structure



Positive Chirp Excitation Experiment

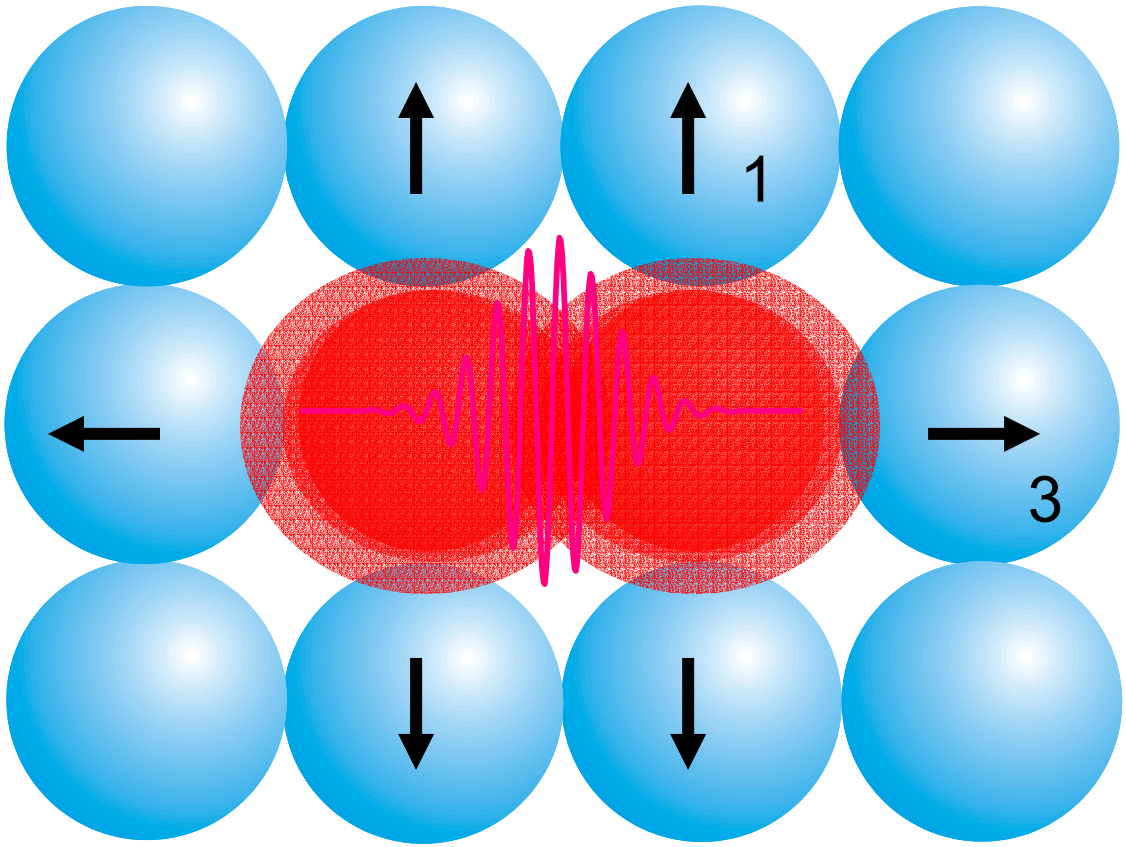


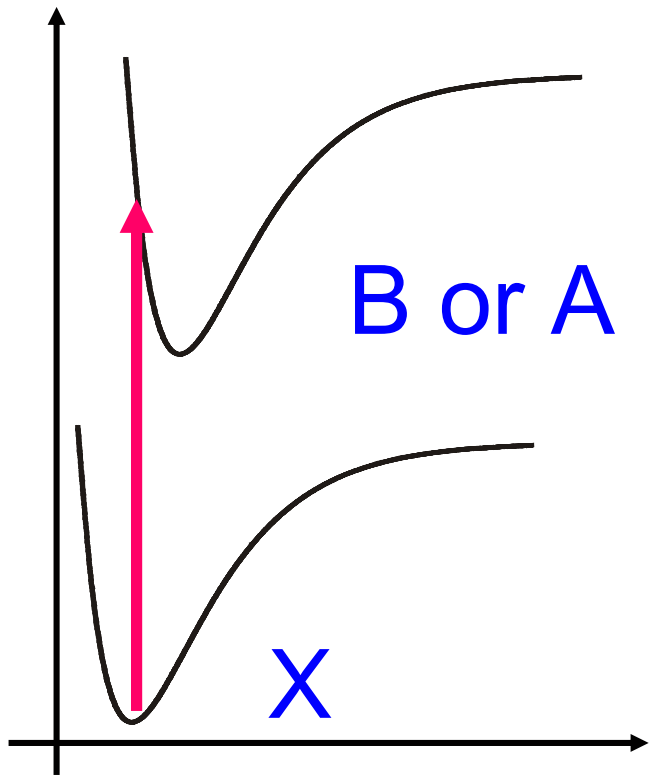


Four level coherence about 1.2 ps

Gühr, Ibrahim, Schwentner, PCCP **6**, 5353-5361 (2004)

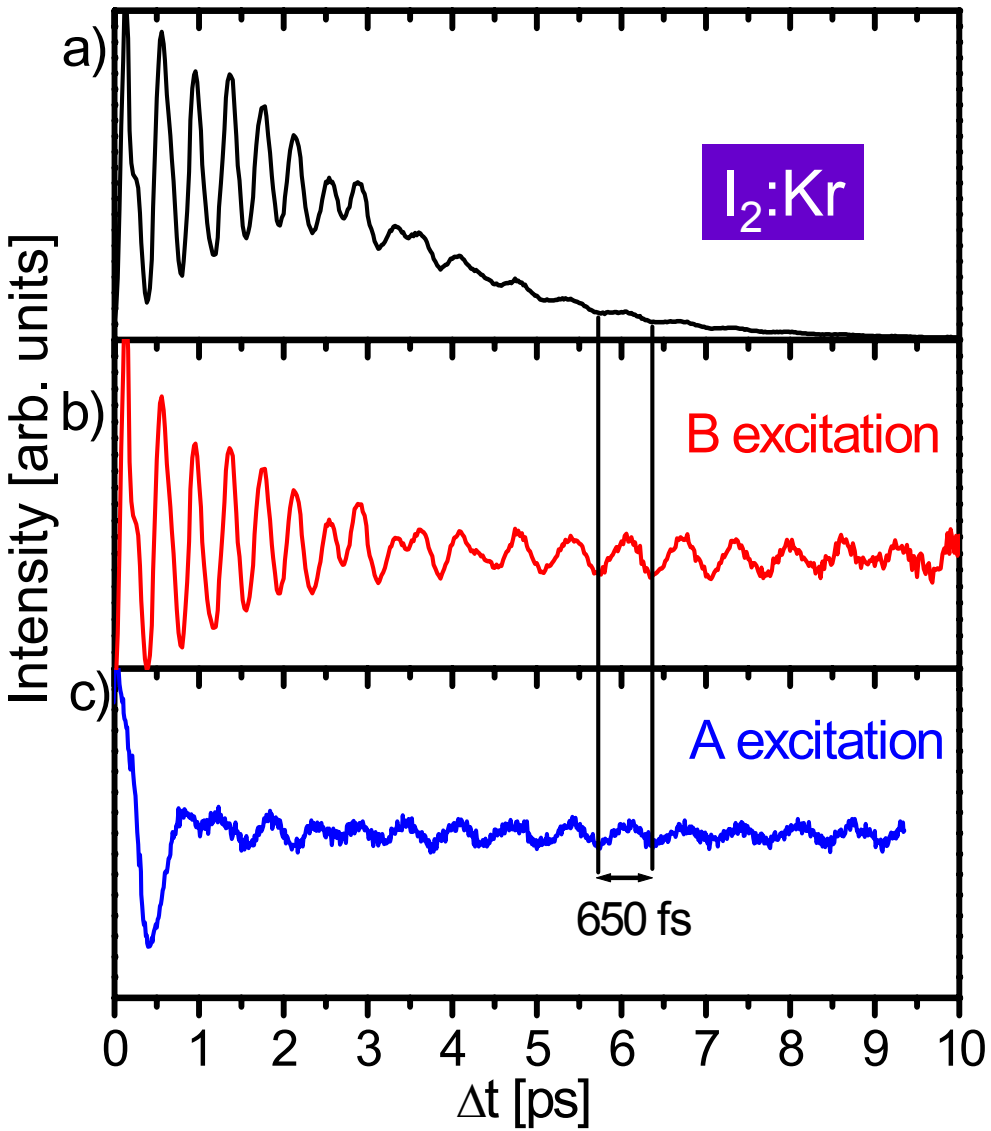
Phonon Excitation





X —Pump Pulse→ B or A

Coherent host dynamics



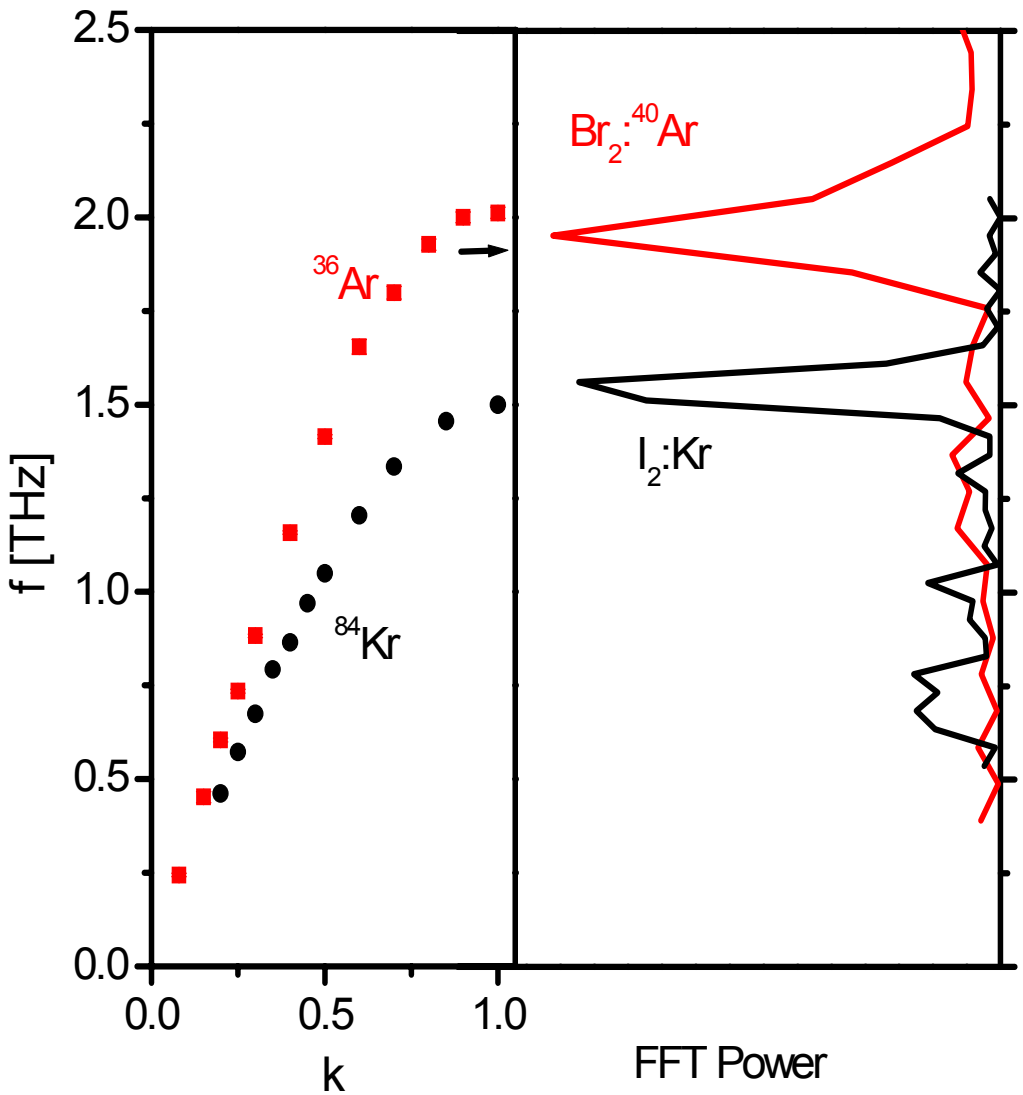
Host Oscillation

- For I_2:Kr: 650 fs (1.5 THz)
- For Br_2:Ar: 500 fs (2 THz)
- Frequency and phase stability
- No forced excitation
- Impulsive excitation
- Decoupling from molec. motion

Gühr, Bargheer, Schwentner, Phys. Rev. Lett. **91**, 085504 (2003)

Gühr, Schwentner, PCCP **7**,760-767 (2005)

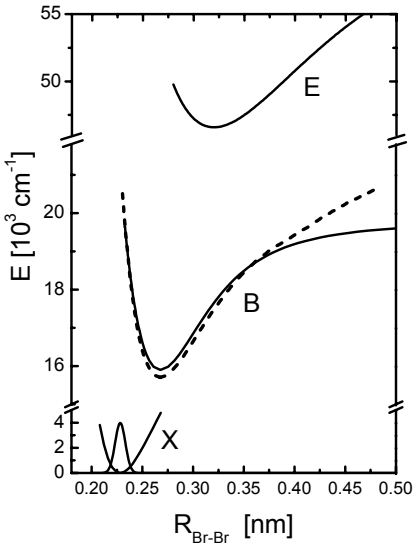
Zone Boundary Phonon



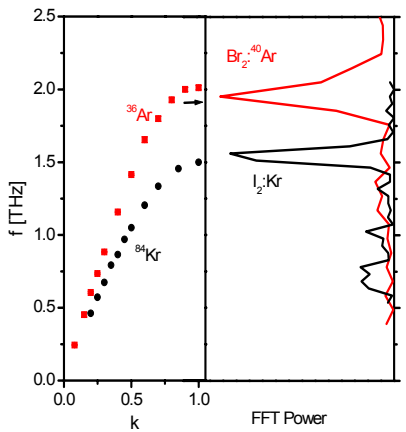
- Host modulation has frequency of ZBP for Ar or Kr crystal
- ZBP has v_g=0, stays at molecule after excitation

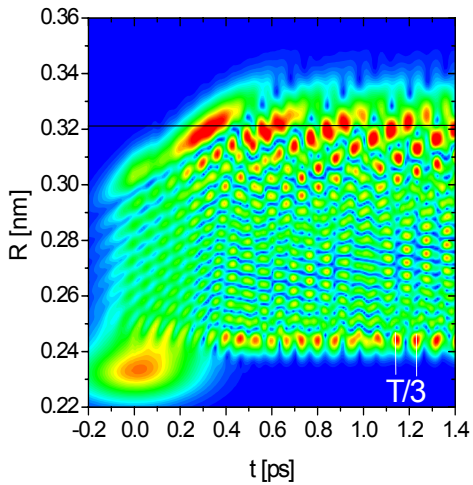
Summary

- Experimental RKR potential



- Coherent control of 1/6 revival: four level coherence





- Coherent ZBP

Thanks to:

- Prof. Schwentner and group (FU Physics)
- Prof. Tannor (Weizmann, Israel)
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- Mr. Zimmermann (glassware)
- Sfb 450 *Analysis and control of ultrafast photoinduced reactions*