



UNIVERSITÀ
DEGLI STUDI DI TRIESTE



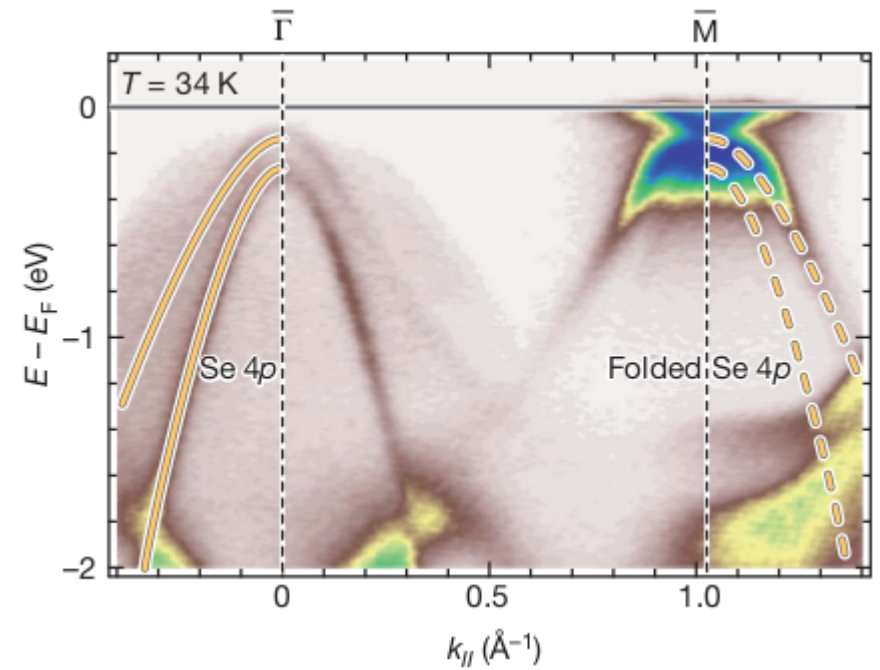
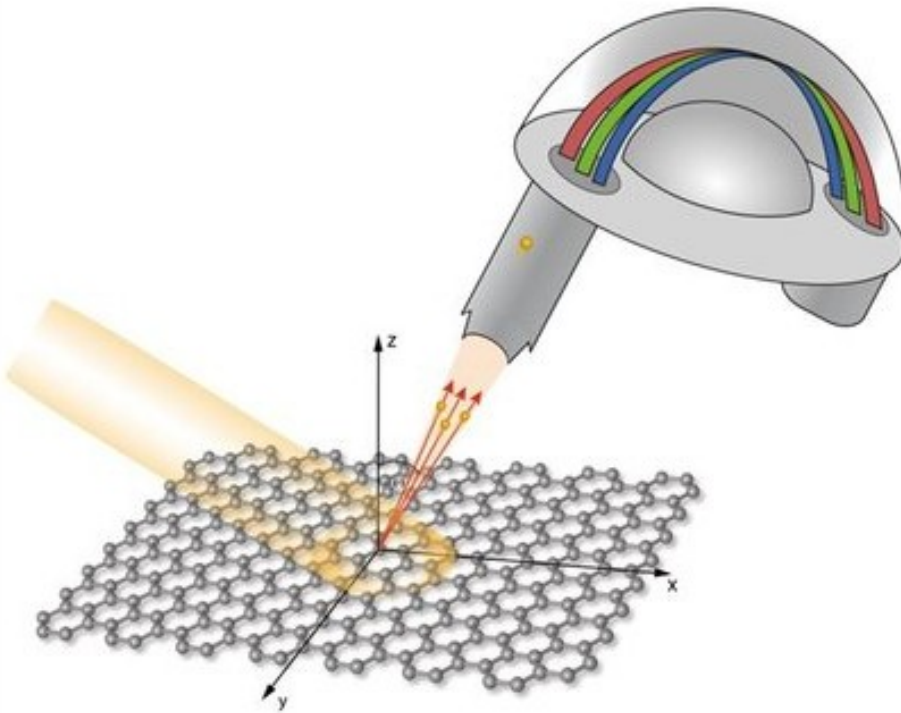
Bypassing the energy-time uncertainty in time-resolved photoemission

Francesco Randi

NGSCES 2016, Trieste

arXiv 1604:08511

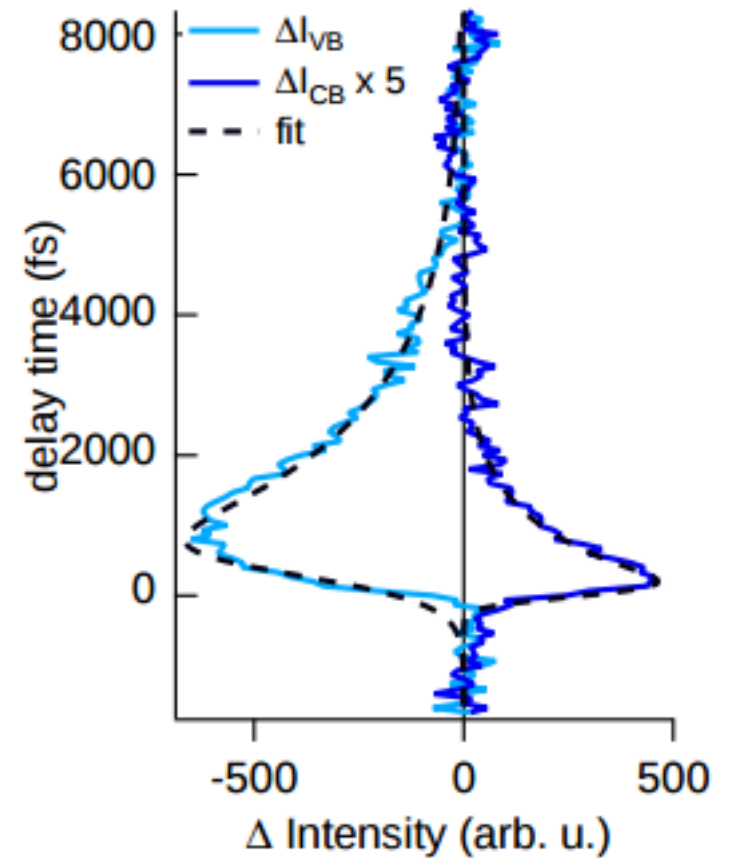
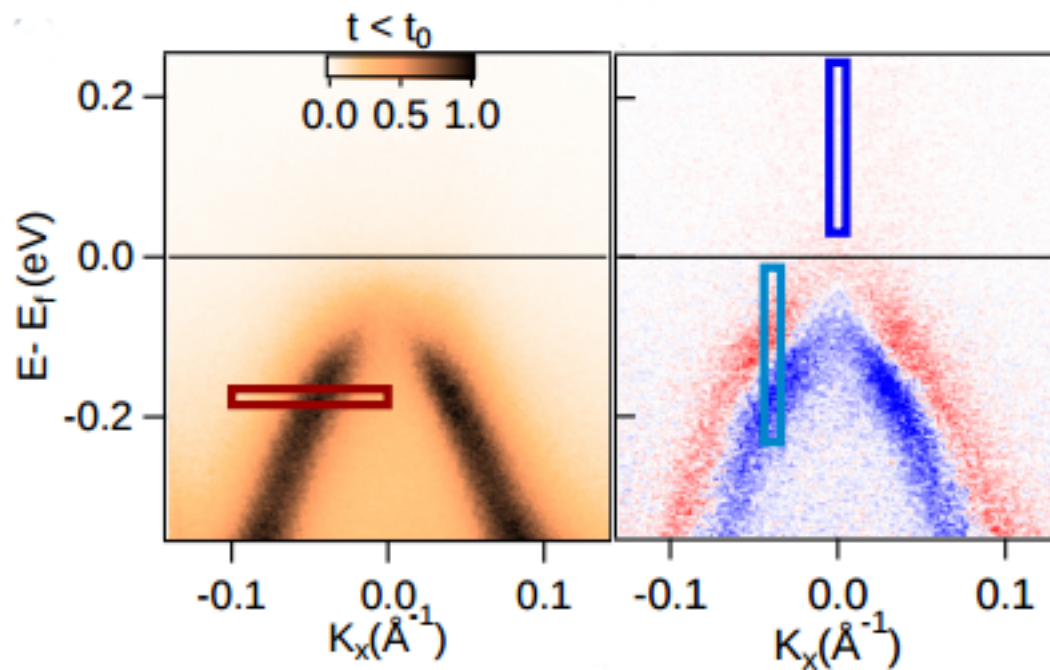
Photoemission



T. Rohwer et al., Nature 471 (2011)

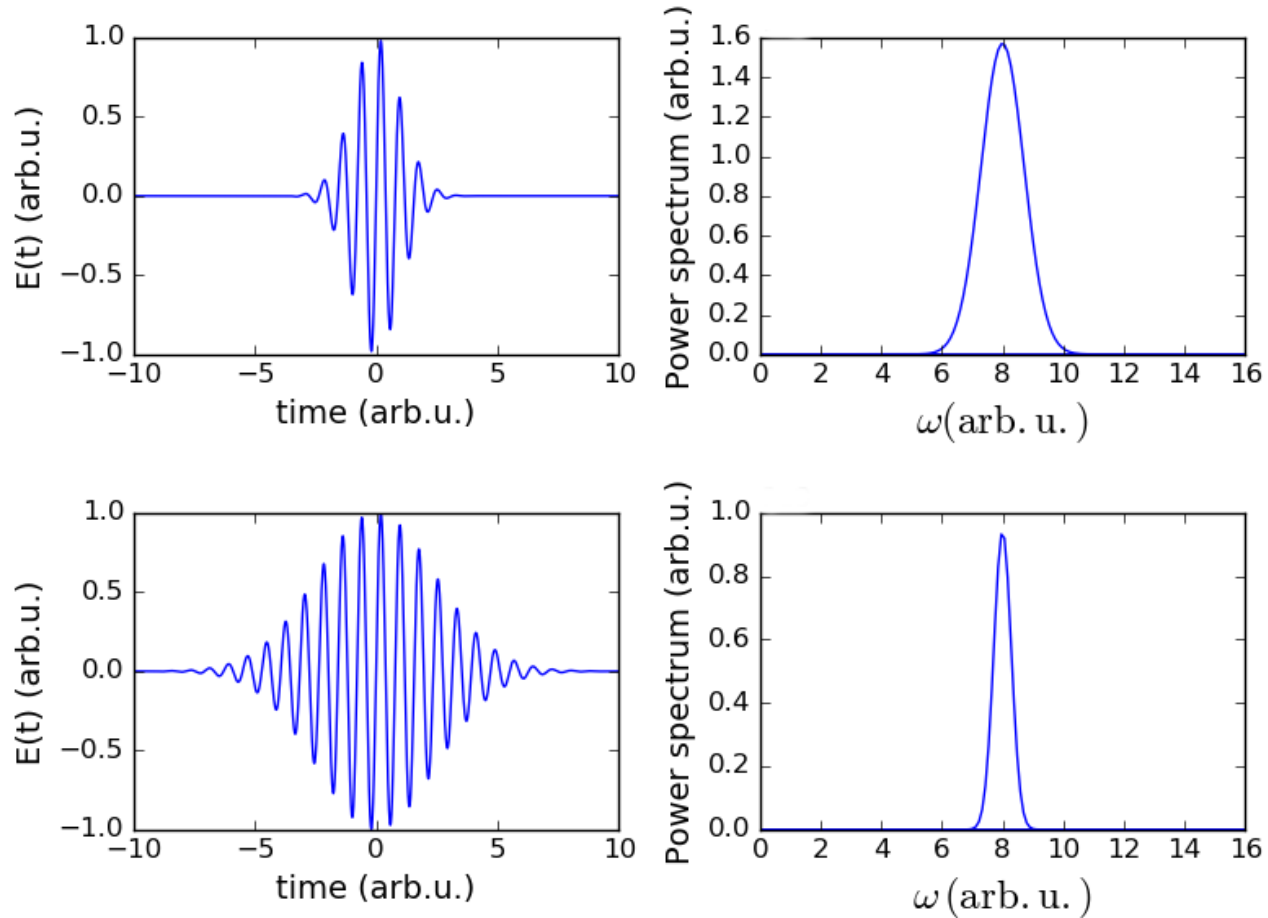
MPSD, MPG

Time-resolved photoemission ^{and angle}

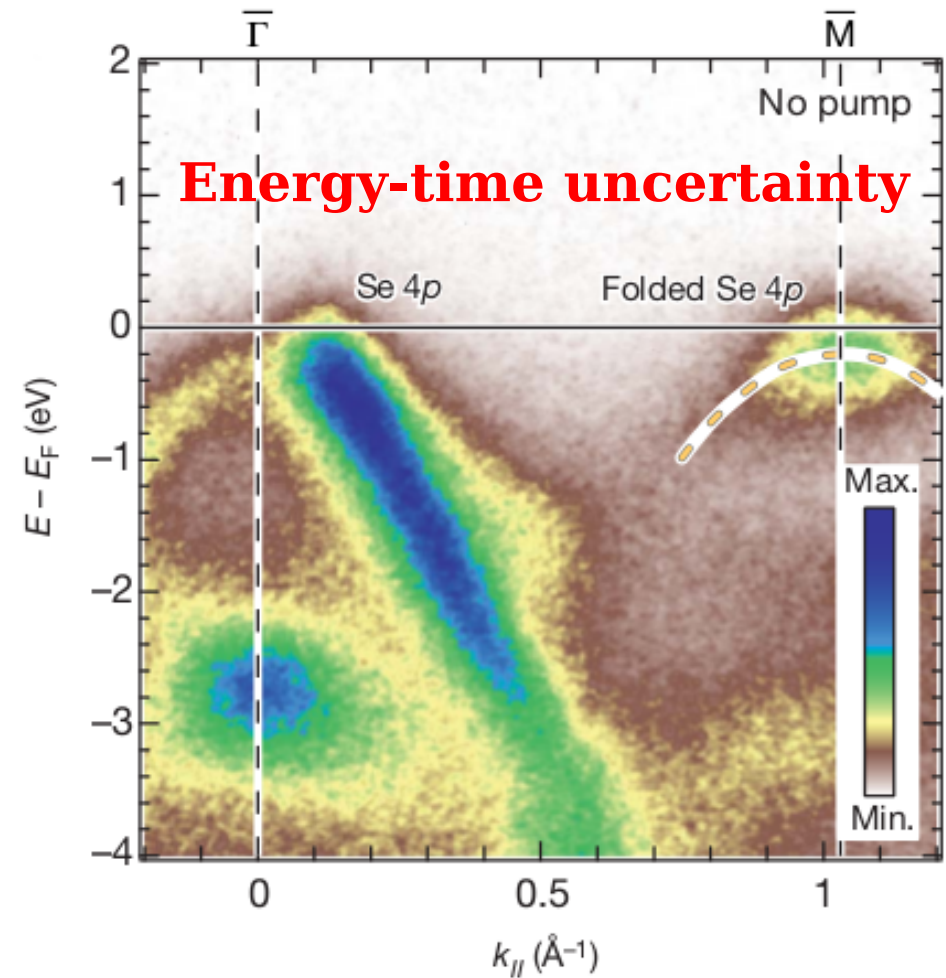
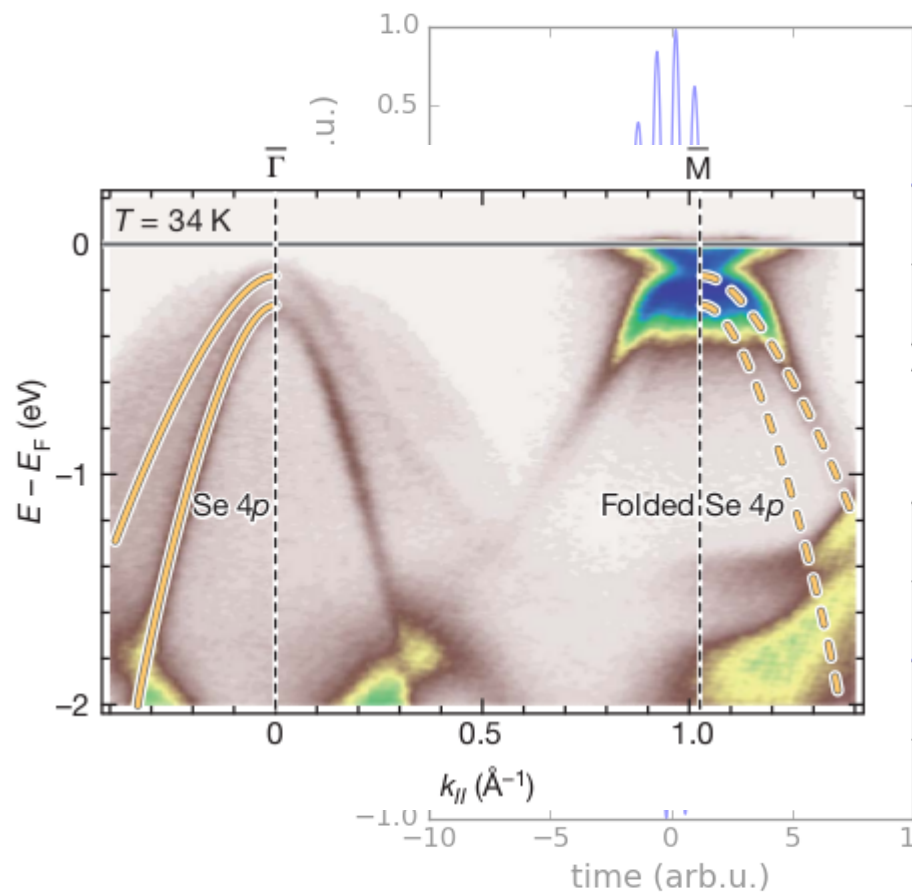


Manzoni et al., PRL 115 (2016)

Time-resolved photoemission ^{and angle}



Time-resolved photoemission ^{and angle}



T. Rohwer et al., Nature 471 (2011)

When does this constitute a serious
problem?

Emergent properties

Emergent properties of solids often show up on small energy scales.

- Kondo screening

Emergent properties

Emergent properties of solids often show up on small energy scales.

- Kondo screening
- Band splitting and gaps, long range order

Emergent properties

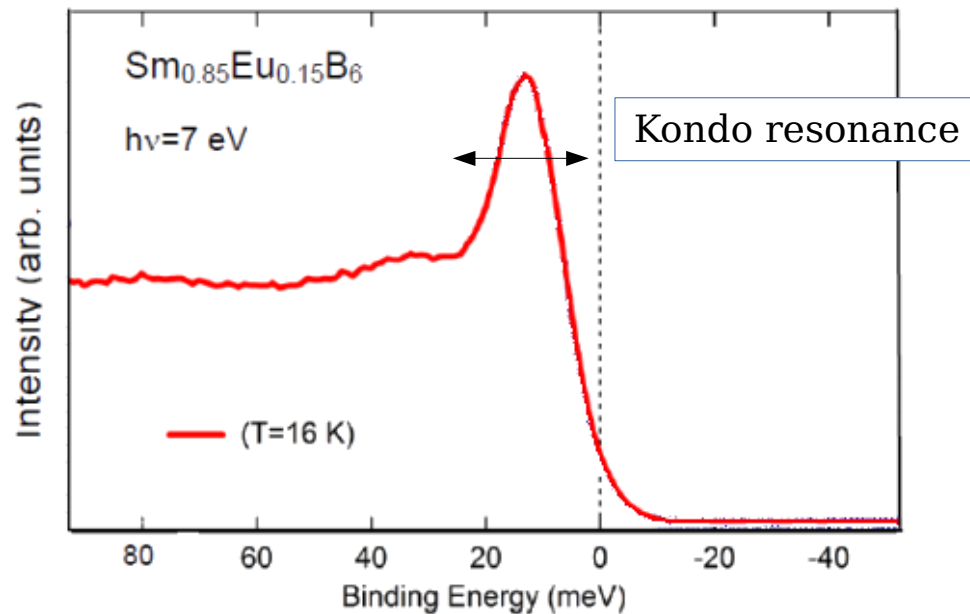
Emergent properties of solids often show up on small energy scales.

- Kondo screening
- Band splitting and gaps, long range order
- Superconducting condensate

Emergent properties

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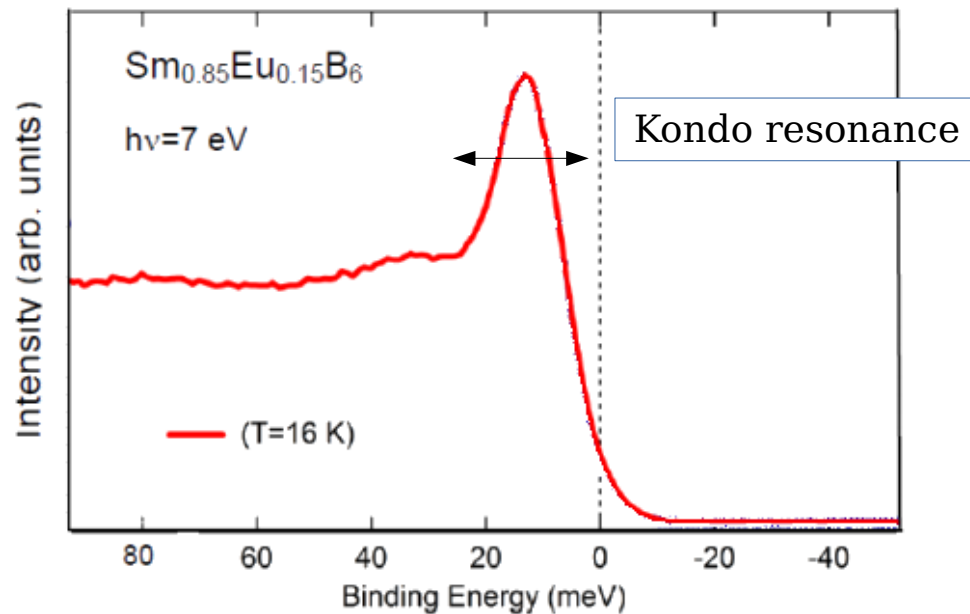


Suga, Imada JPSJ 83 (2014)

Emergent properties

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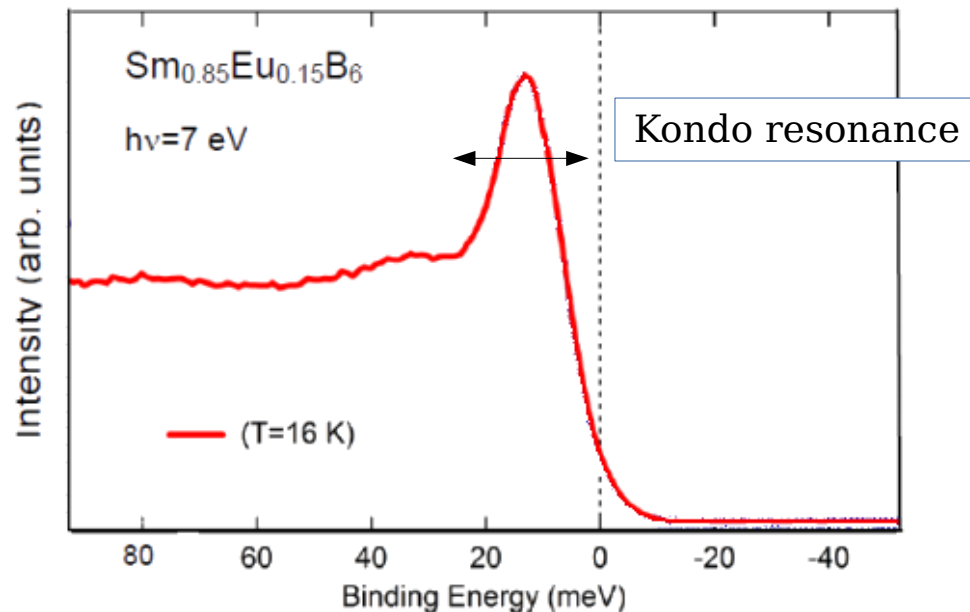
Can we observe its dynamics on timescales *beyond the spectral uncertainty limit?*

$$\leq \frac{\hbar}{\Delta E} \quad ?$$

Emergent properties

Emergent properties of solids often show up on small energy scales.

- Kondo screening
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Can we observe its dynamics on timescales *beyond the spectral uncertainty limit?*

$$\leq \frac{\hbar}{\Delta E} \quad ?$$

Timescale for the screening can be faster than the inverse width of the spectral feature

Lechtenberg PRB 90, 2014

Suga, Imada JPSJ 83 (2014)

Is the energy-time uncertainty a
fundamental limitation?

Outline

- Standard time-resolved photoemission
- How to bypass the energy-time uncertainty
- Example: the buildup of the Kondo resonance

Time-resolved photoemission

From perturbation theory...

Time-resolved photoemission

From perturbation theory...

$$I(E, t_p) \propto \int dt dt' e^{-iE(t-t')} G^<(t_p + t, t_p - t') S(t, t')$$

Time-resolved photoemission

From perturbation theory...

$$I(E, t_p) \propto \int dt dt' e^{-iE(t-t')} \underbrace{G^<(t_p + t, t_p - t') S(t, t')}_{\text{Properties of the solid}}$$

Properties of the solid
Contains the true timescales

Time-resolved photoemission

From perturbation theory...

$$I(E, t_p) \propto \int dt dt' e^{-iE(t-t')} \underbrace{G^<(t_p + t, t_p - t')}_{\text{Properties of the solid}} \underbrace{S(t, t')}_{\text{probe pulse envelope}}$$

Properties of the solid

$$S(t, t') = s(t) s(t')^*$$

$s(t)$ probe pulse envelope

Time-resolved photoemission

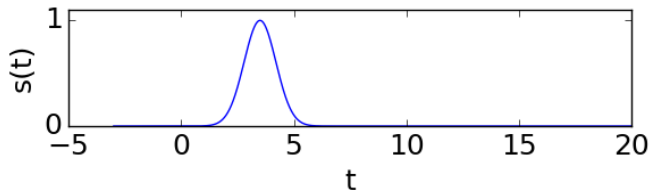
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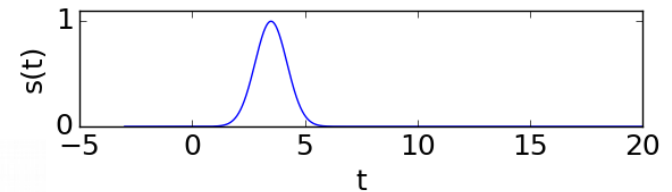
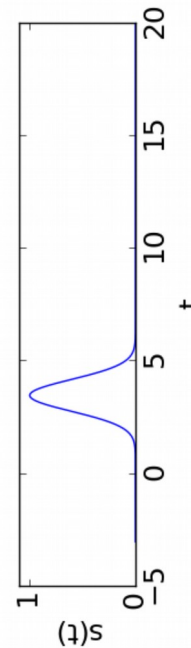
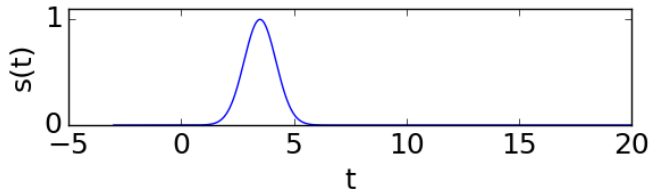
Time-resolved photoemission

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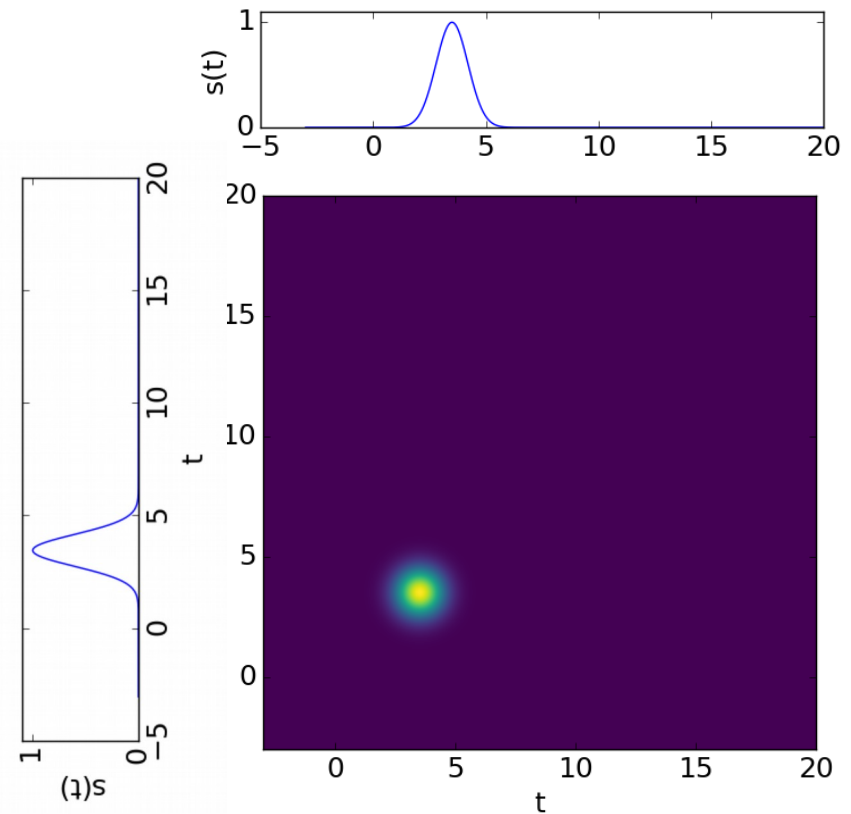
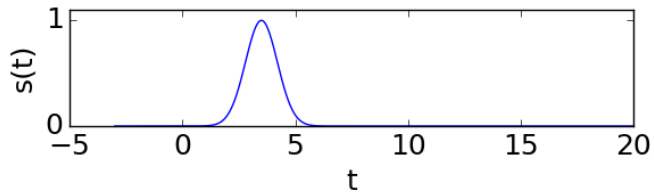
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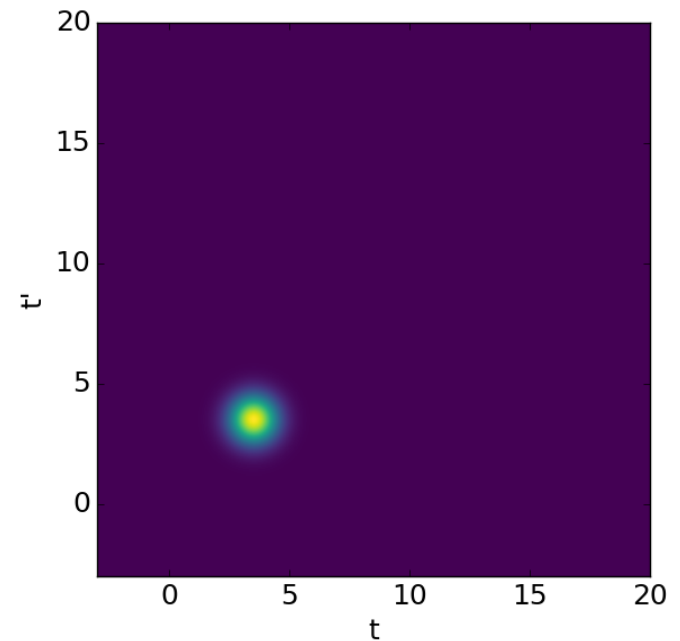
$s(t)$ probe pulse envelope



Time-resolved photoemission

From perturbation theory...

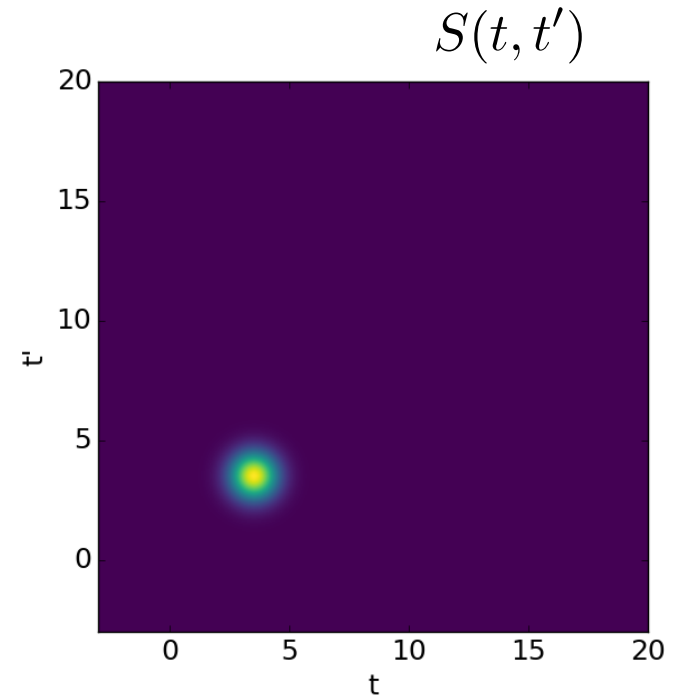
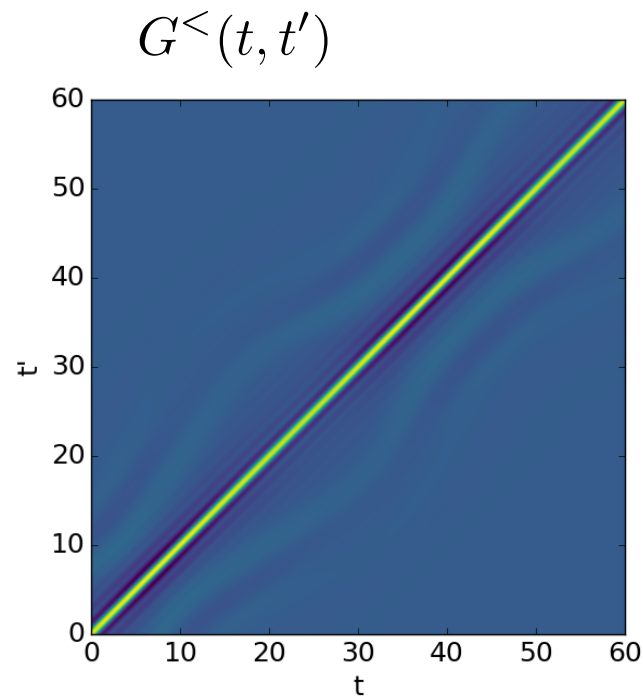
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Time-resolved photoemission

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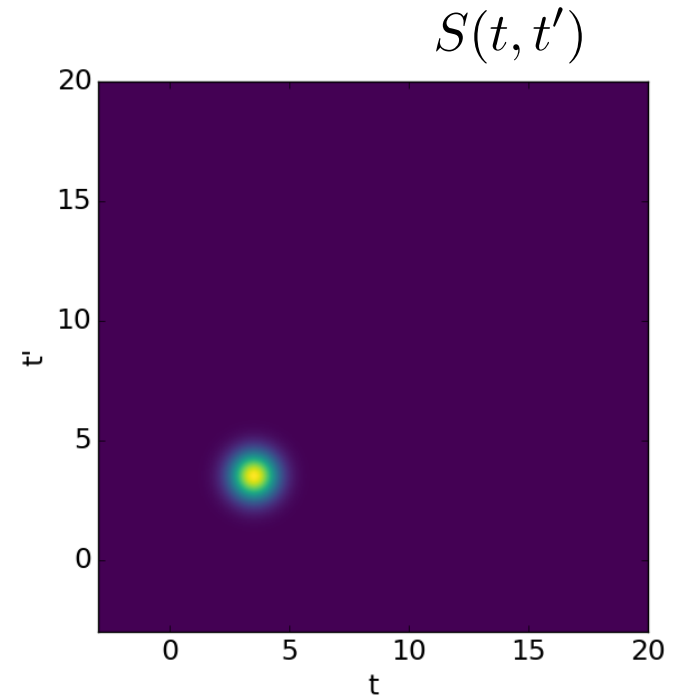
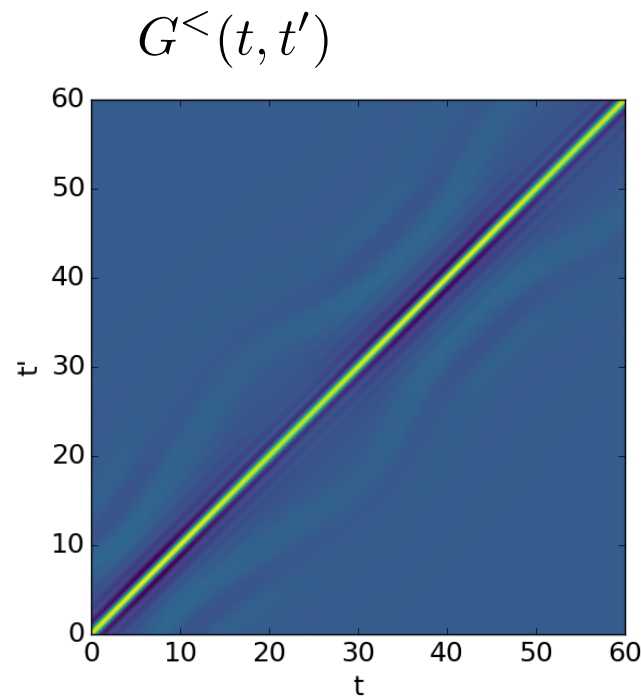
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Time-resolved photoemission

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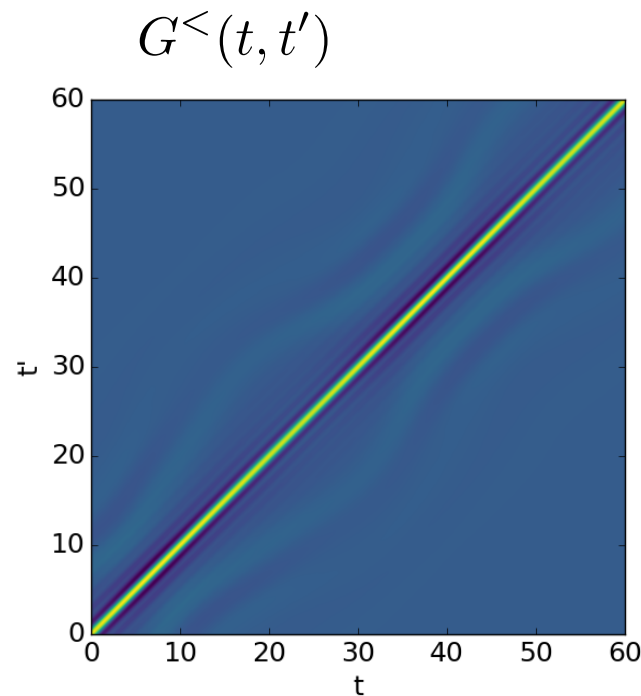


Filtering with $S(t, t')$ + Fourier transform on the antidiagonal

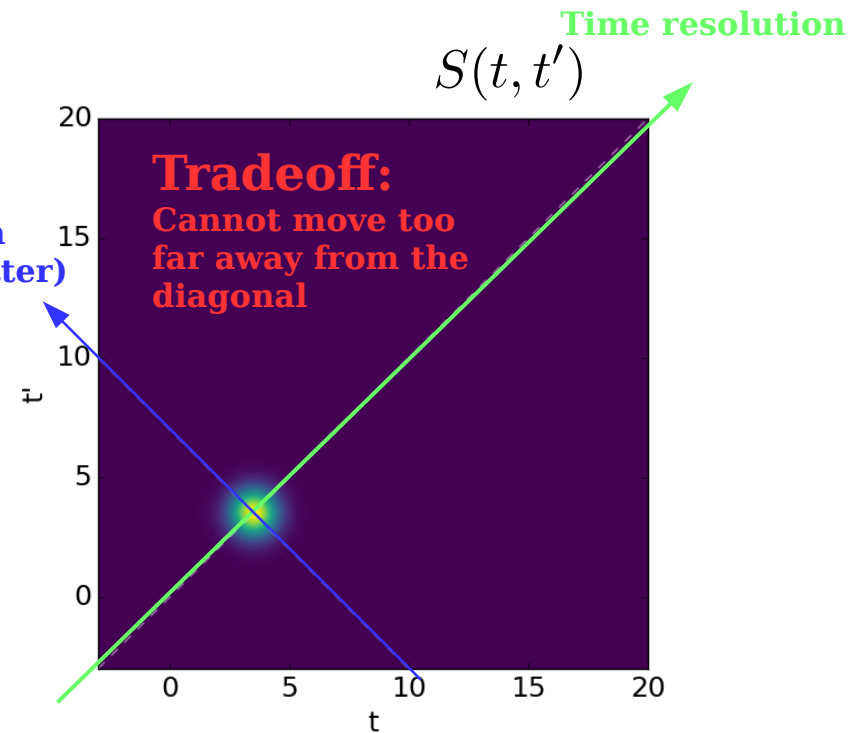
Time-resolved photoemission

From perturbation theory...

$$I(E, t_p) \propto \int dt dt' e^{-iE(t-t')} G^<(t_p + t, t_p - t') S(t, t')$$



Spectral resolution
(the longer the better)

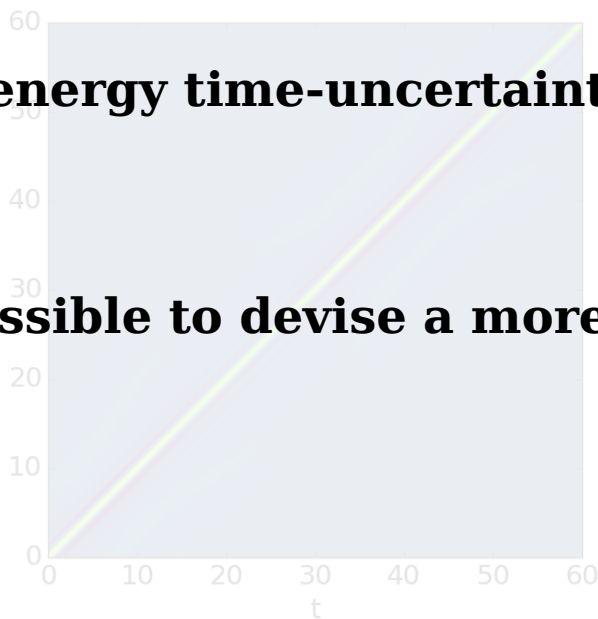


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$G^<(t, t')$

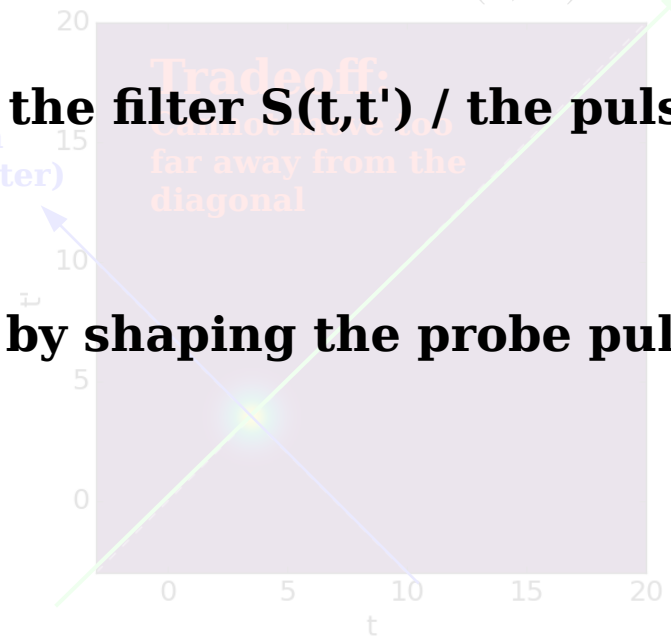


The energy time-uncertainty comes from the filter $S(t, t')$ / the pulse.

Spectral resolution
(the longer the better)

$S(t, t')$

Time resolution

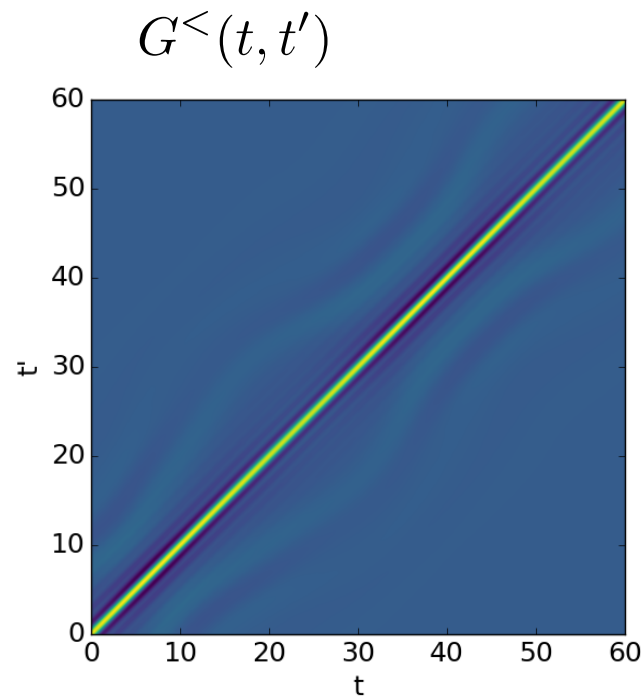


Is it possible to devise a more useful $S(t, t')$ by shaping the probe pulse?

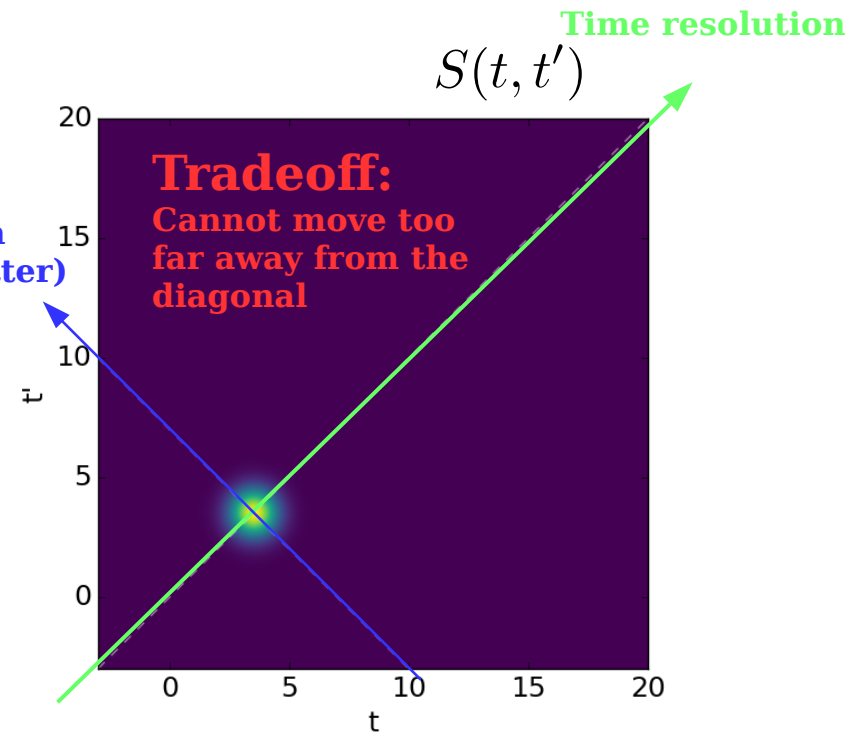
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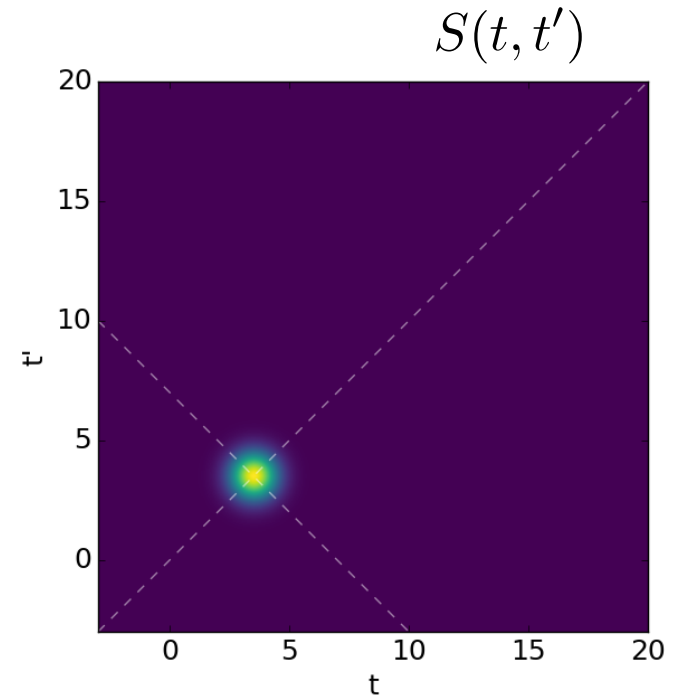
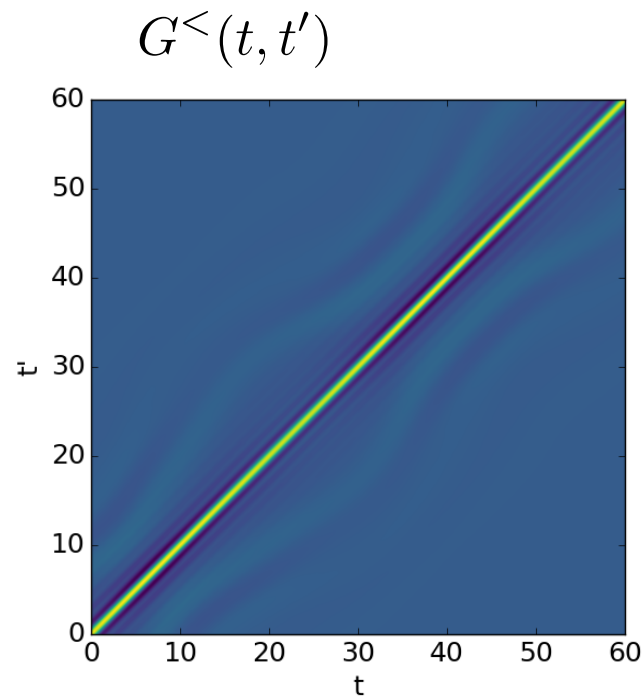


Spectral resolution
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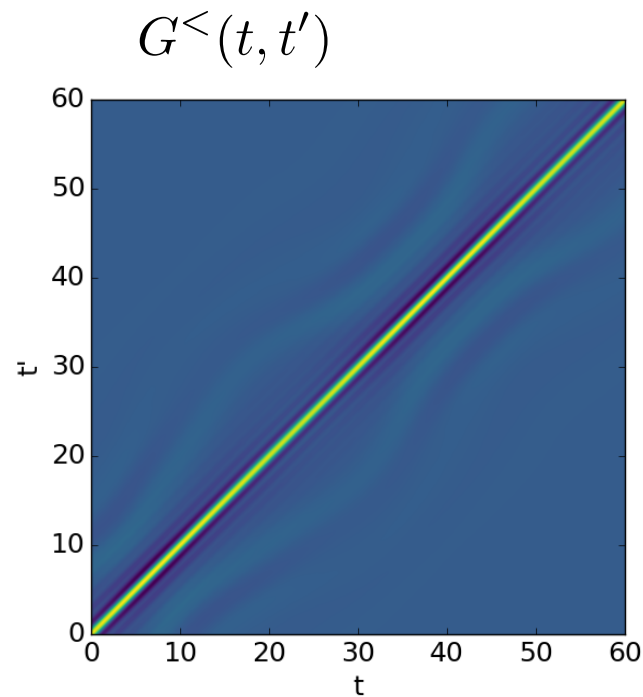


Time-resolved photoemission

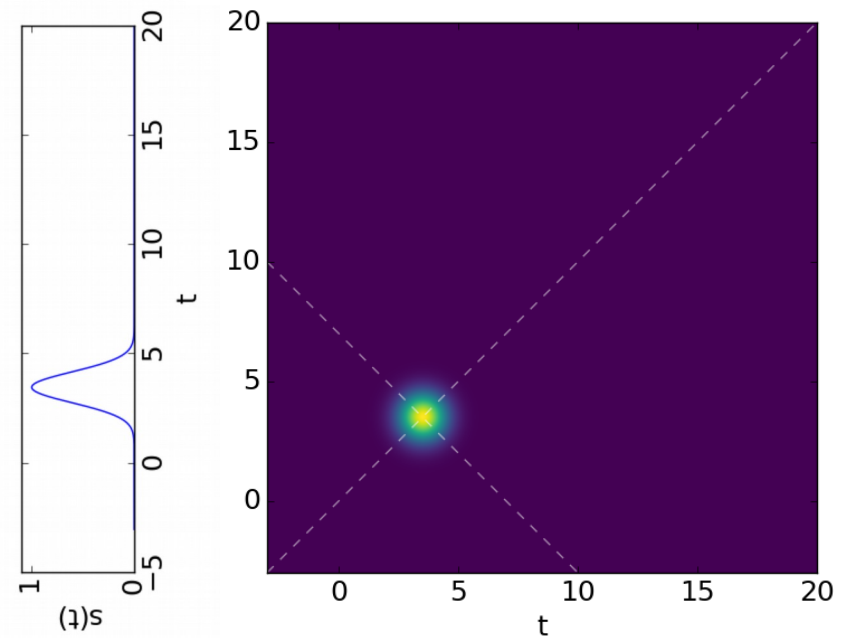
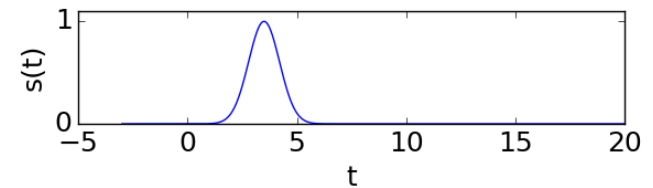
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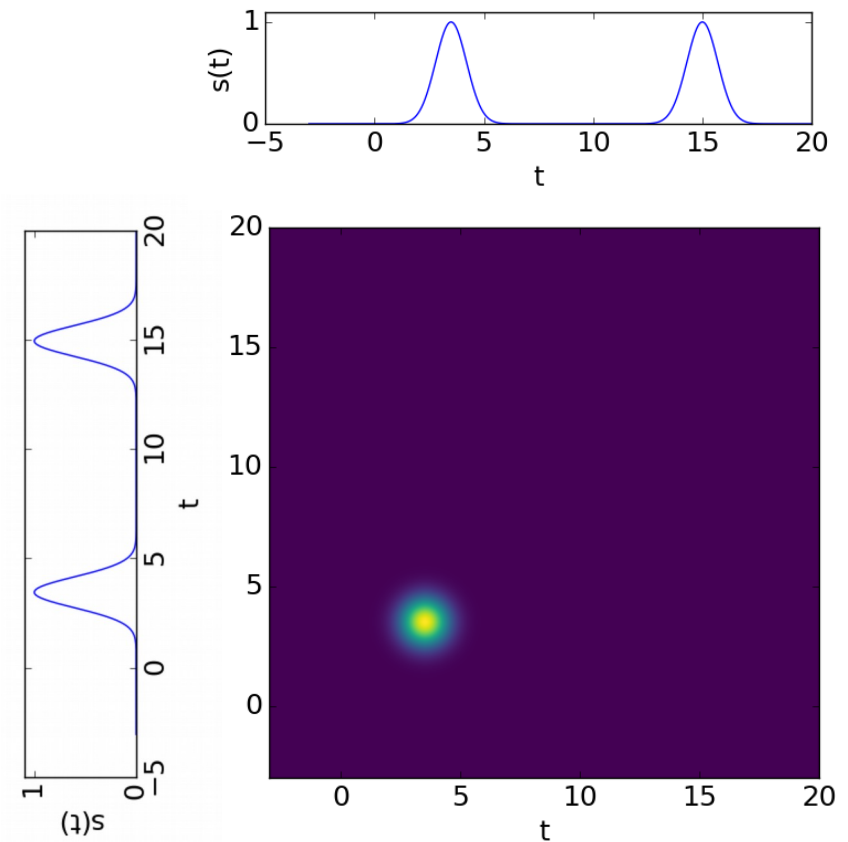
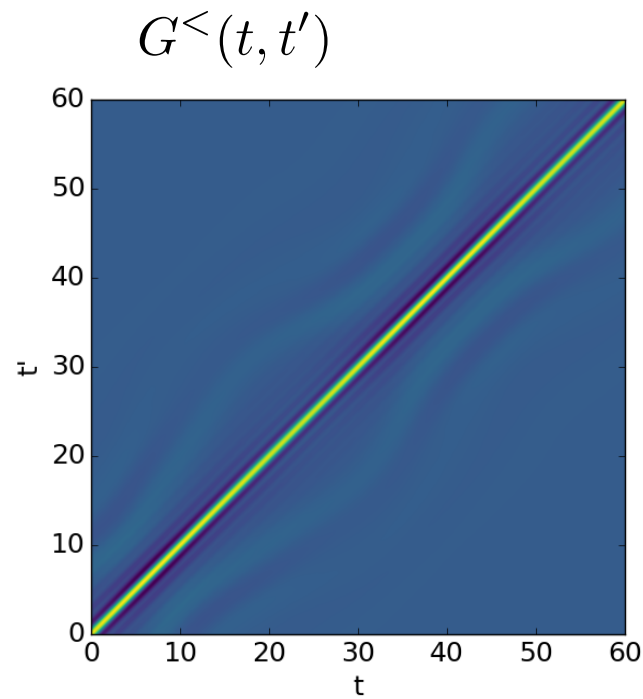
Time-resolved photoemission



$$S(t, t') = s(t)s(t')^*$$

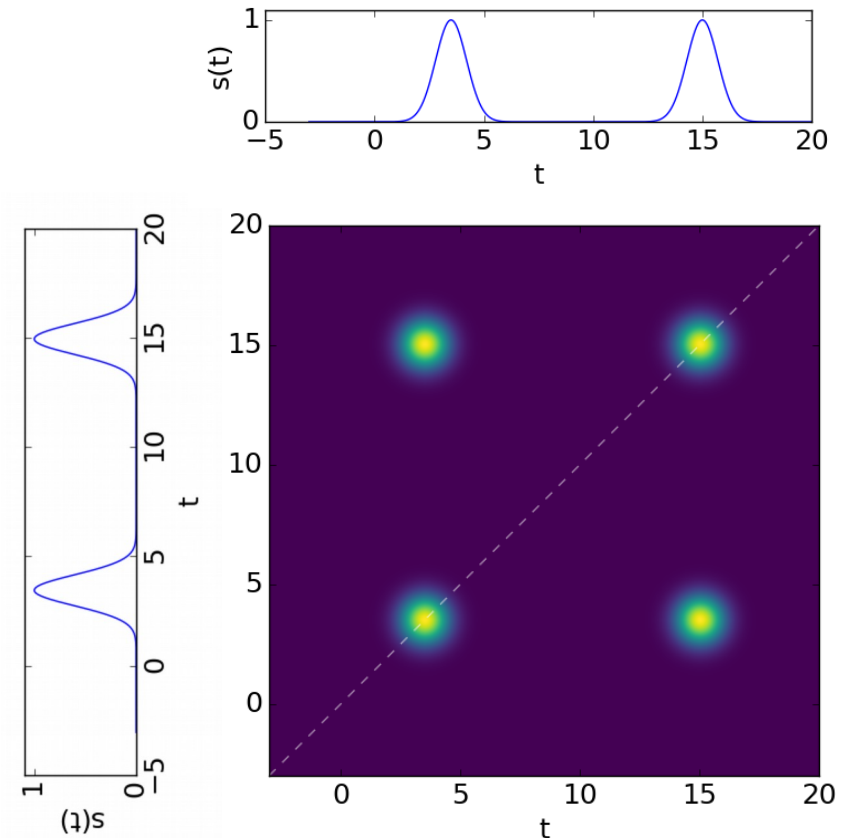
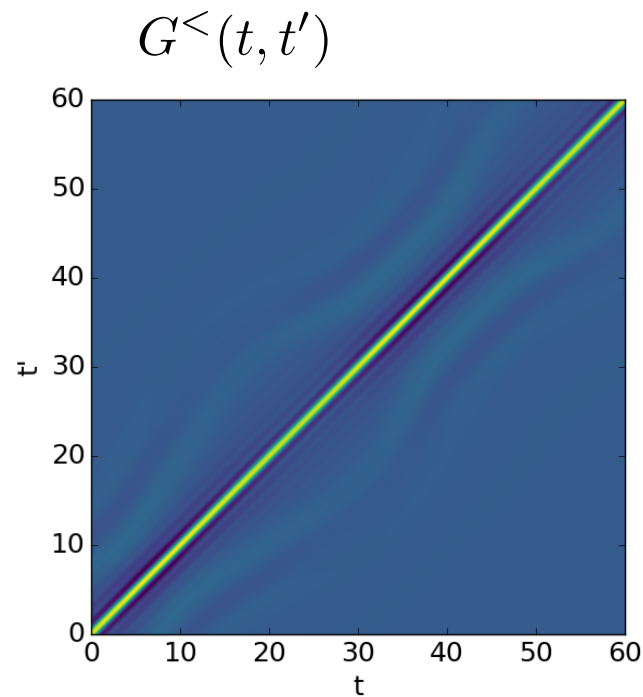


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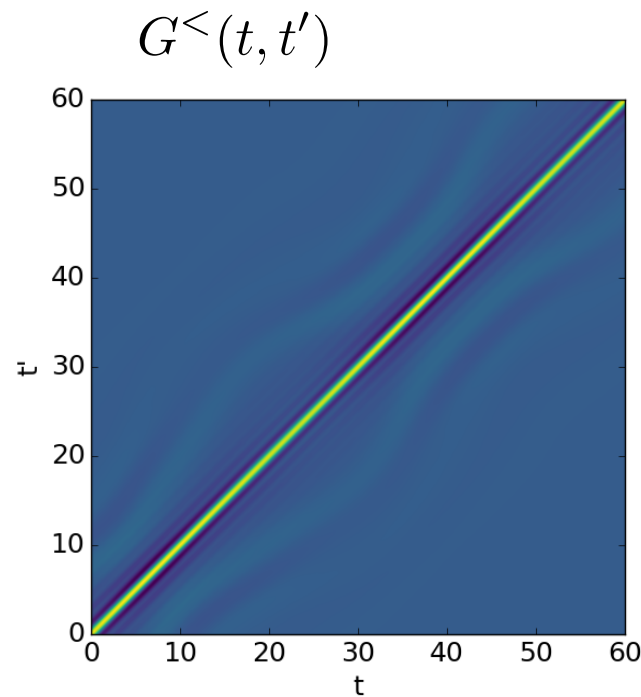
Time-resolved photoemission

“two-dimensional” photoemission

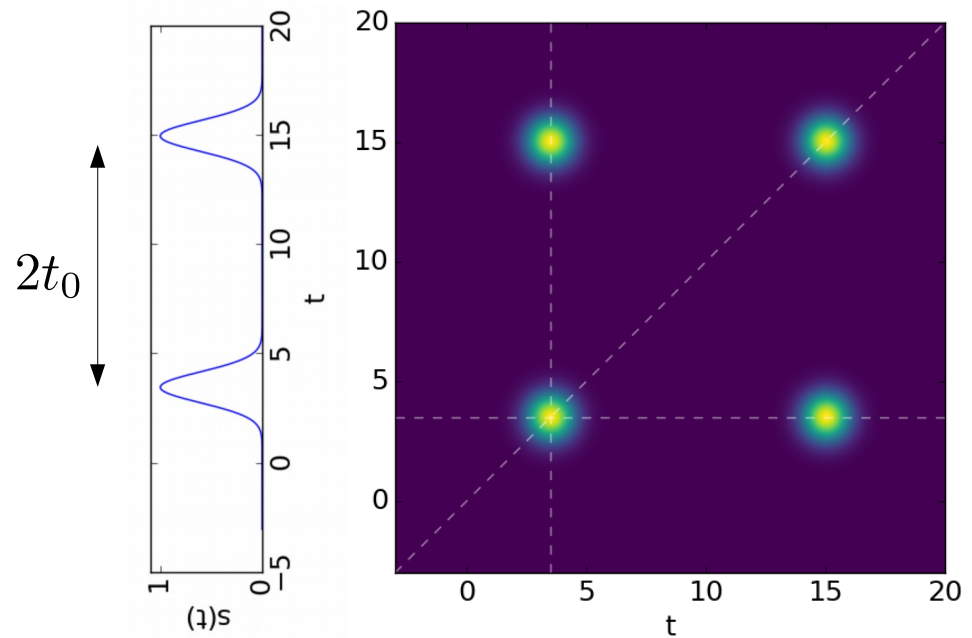
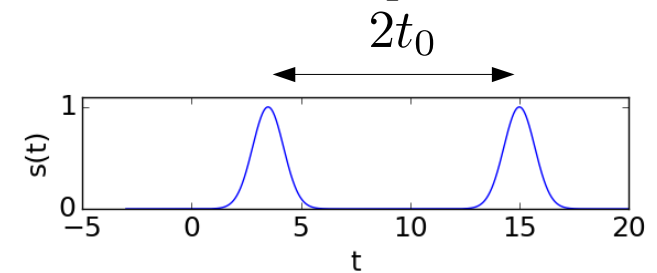


+ fixed phase relation

Time-resolved photoemission



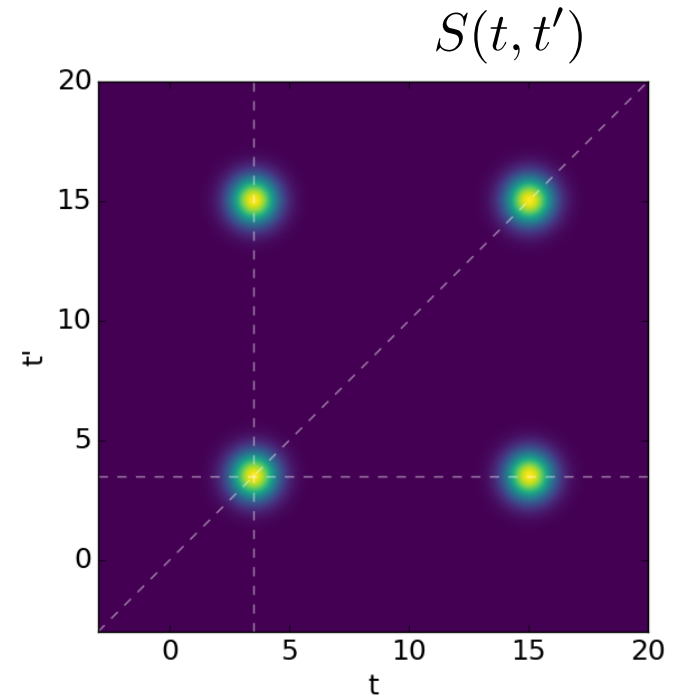
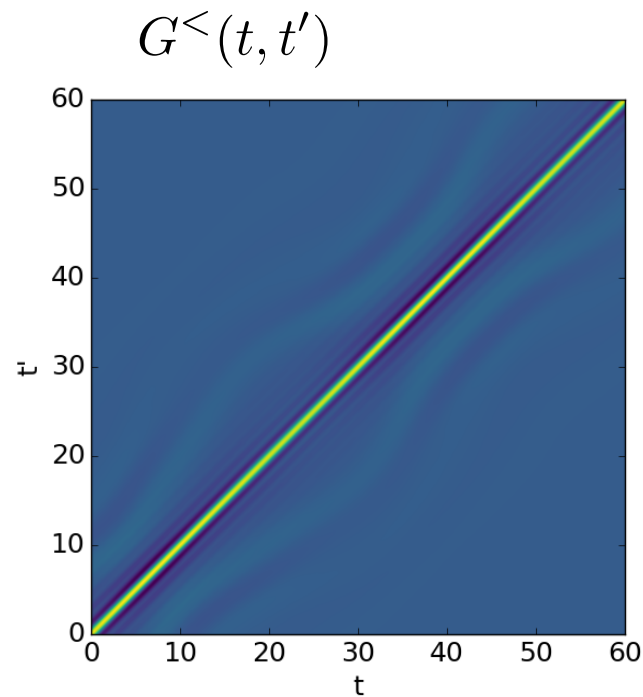
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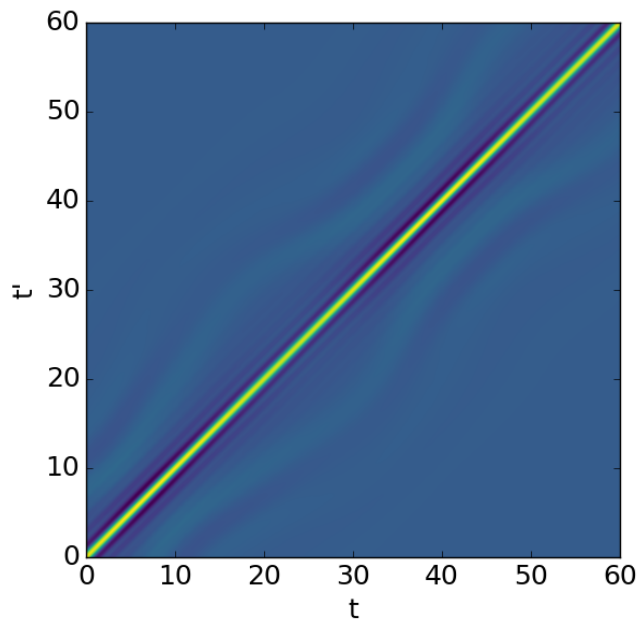
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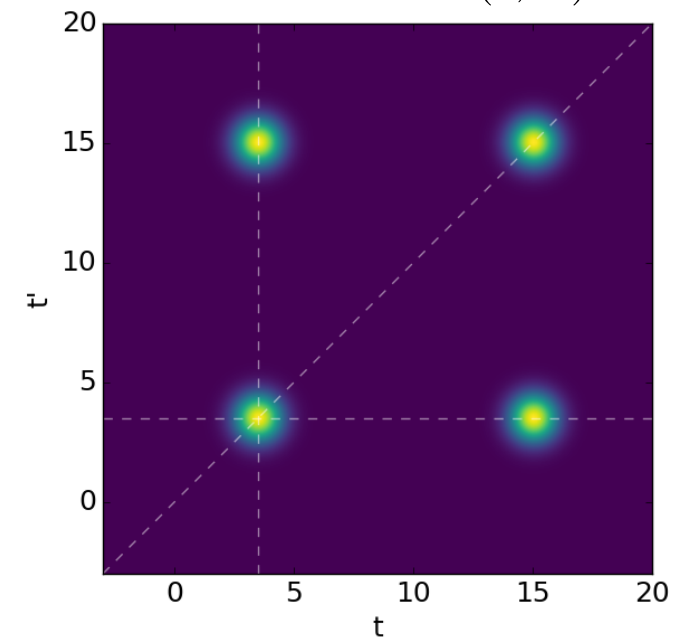
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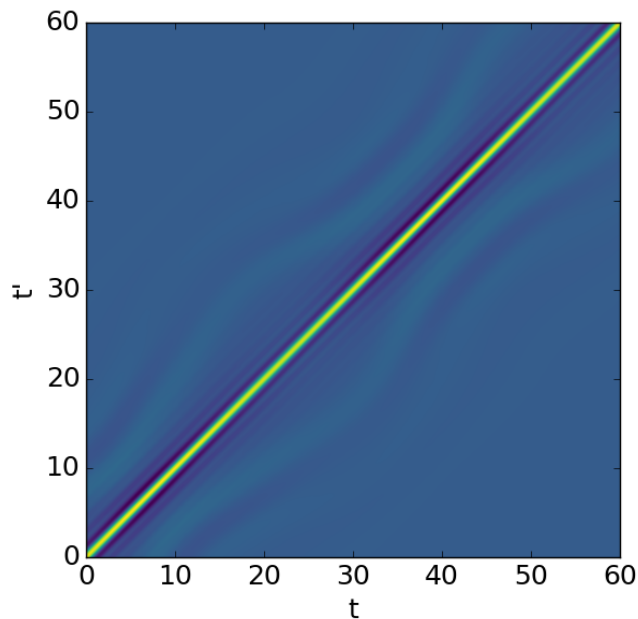
$S(t, t')$



Time-resolved photoemission

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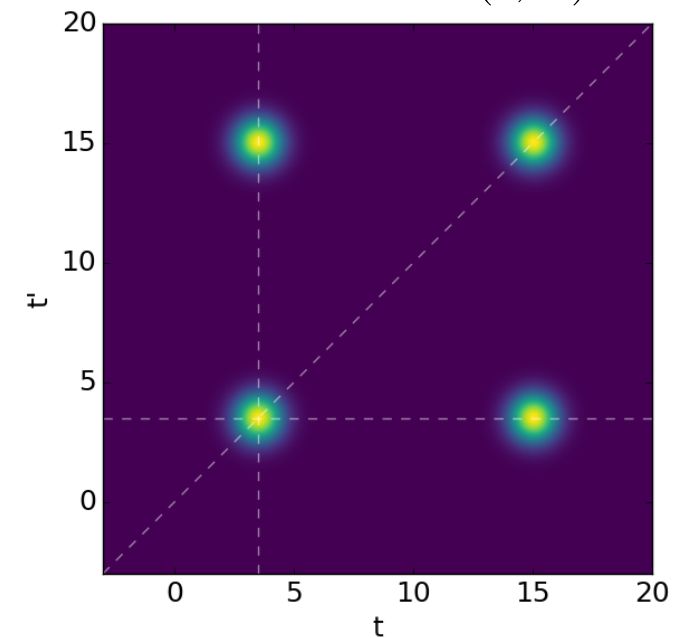
$G^<(t, t')$



4 terms

- 2 on the diagonal can be subtracted from single pulse expt.
- 2 off-diagonal oscillate as a function of photoelectron energy

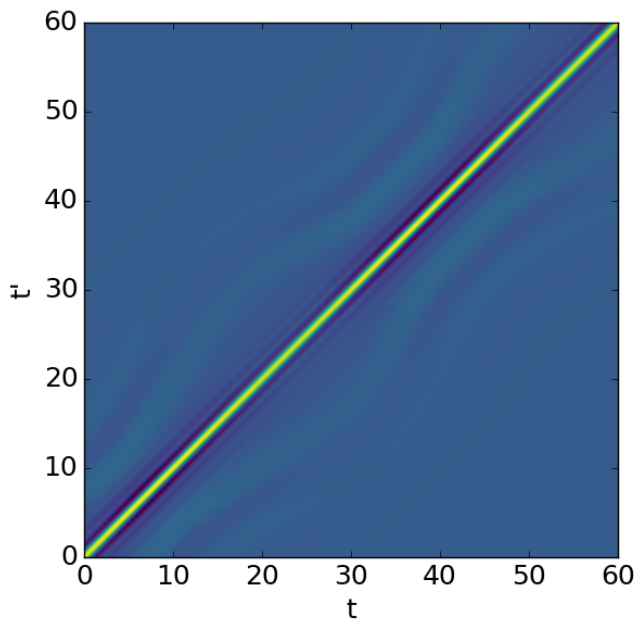
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Time-resolved photoemission

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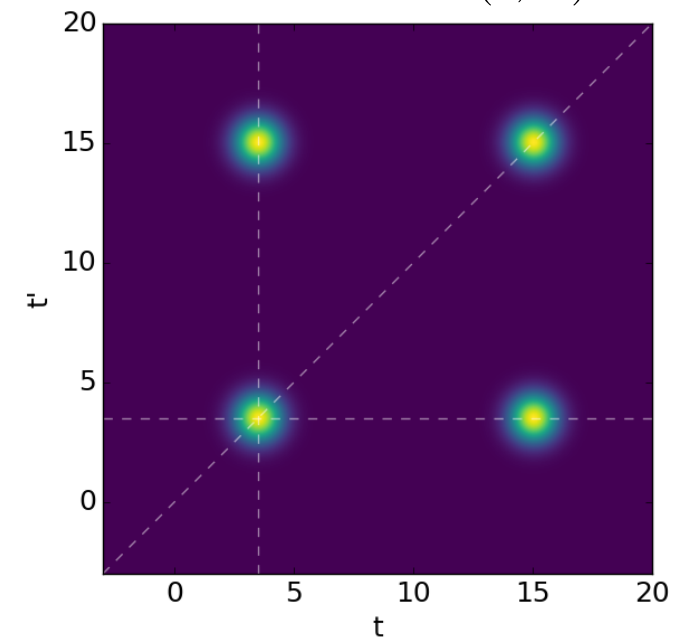
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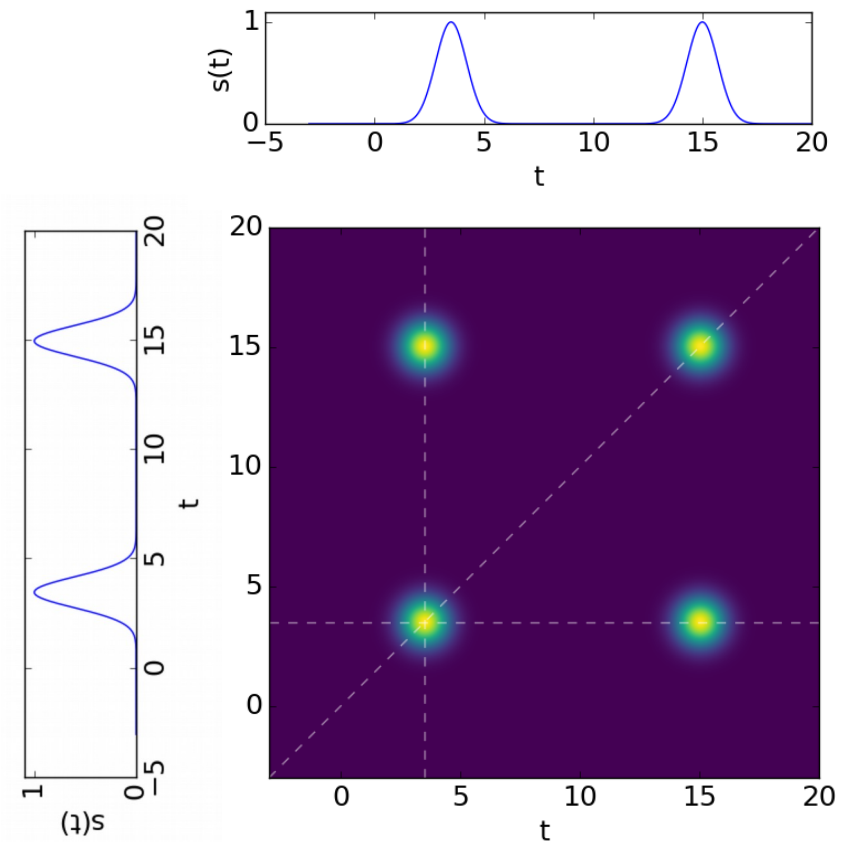


Tomographic (t_p, t_0) reconstruction of the Green's function.

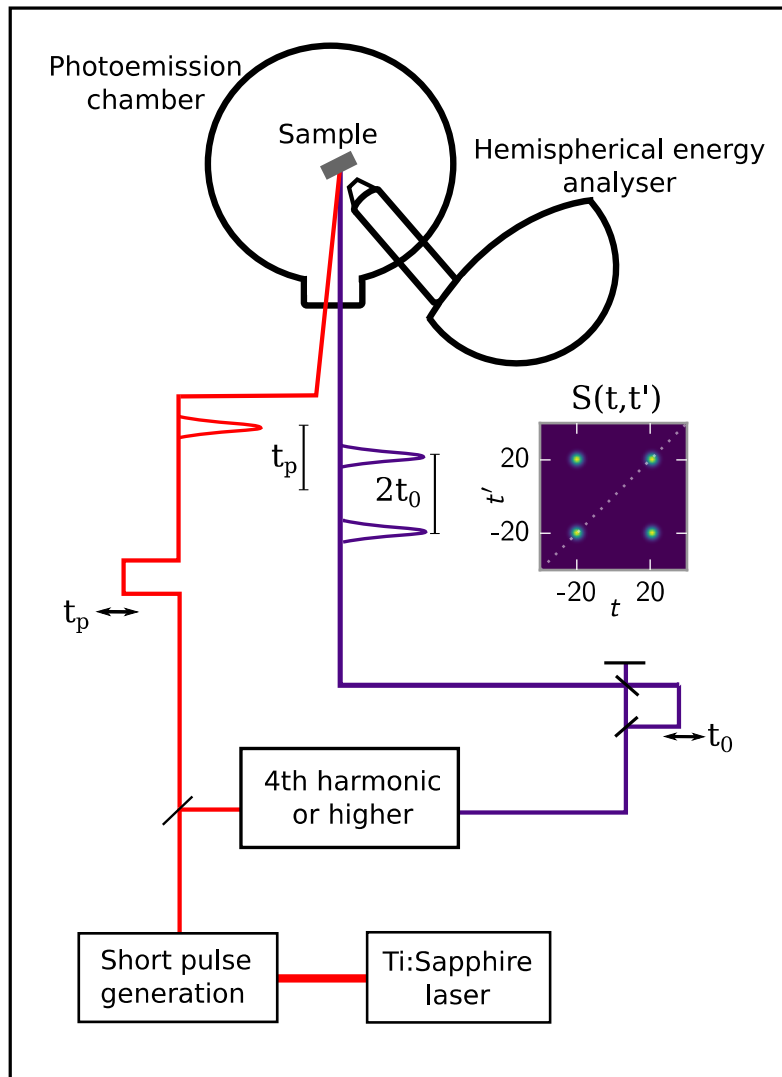
Time-resolved photoemission

“two-dimensional” photoemission

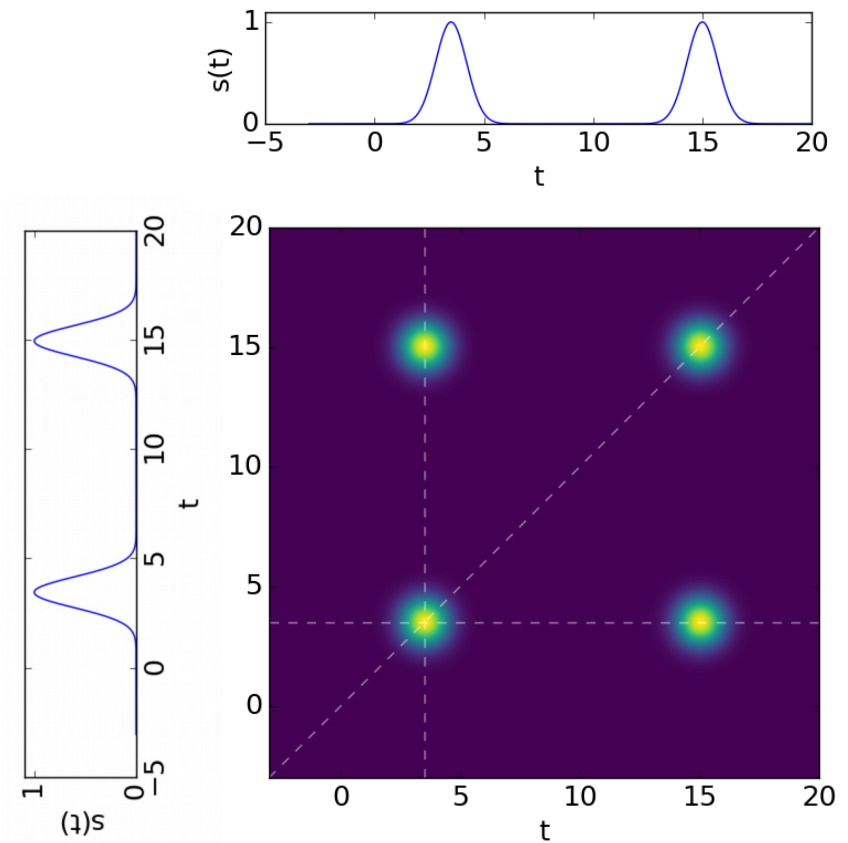
How could this be realized in an experimental set-up?



Time-resolved photoemission



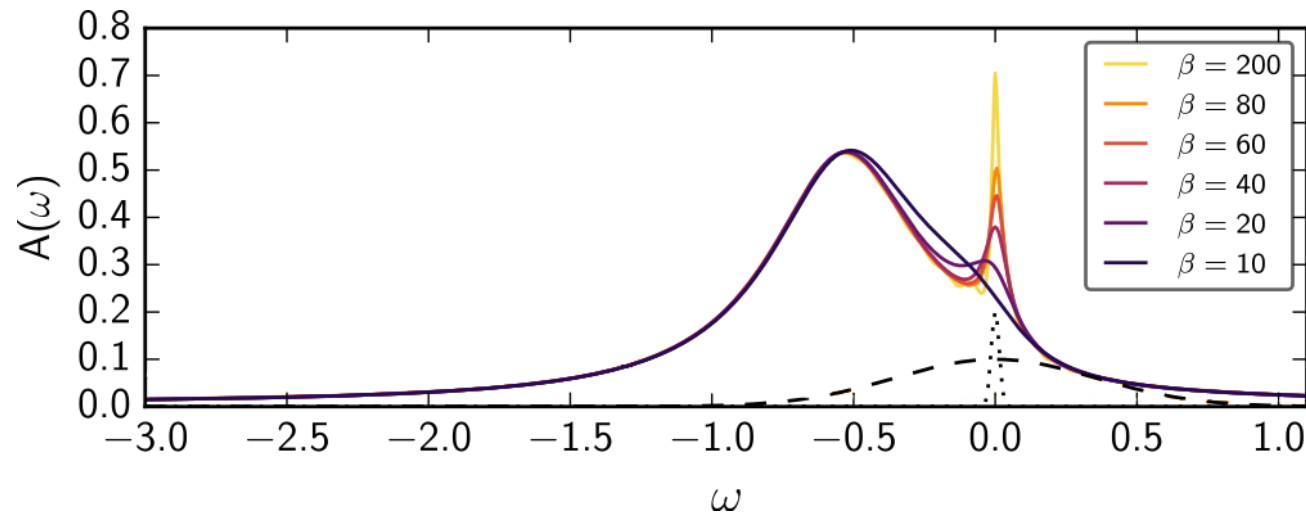
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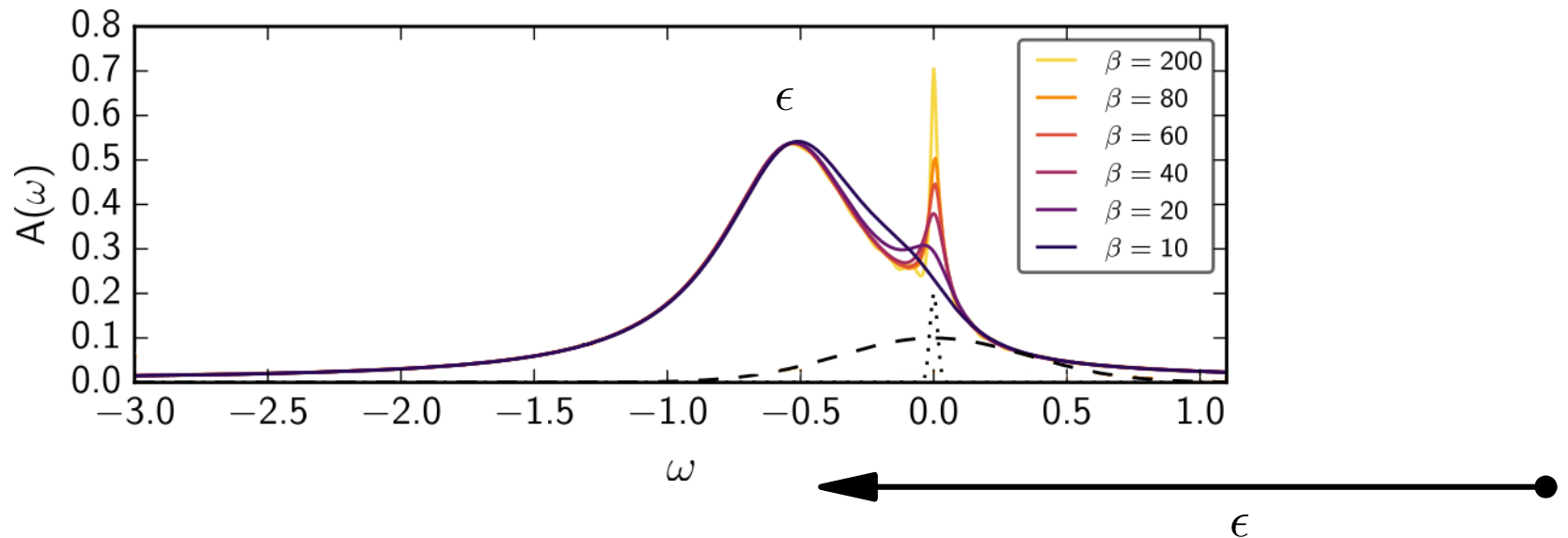
An illustrative example

The buildup of the Kondo peak

Buildup of the Kondo screening

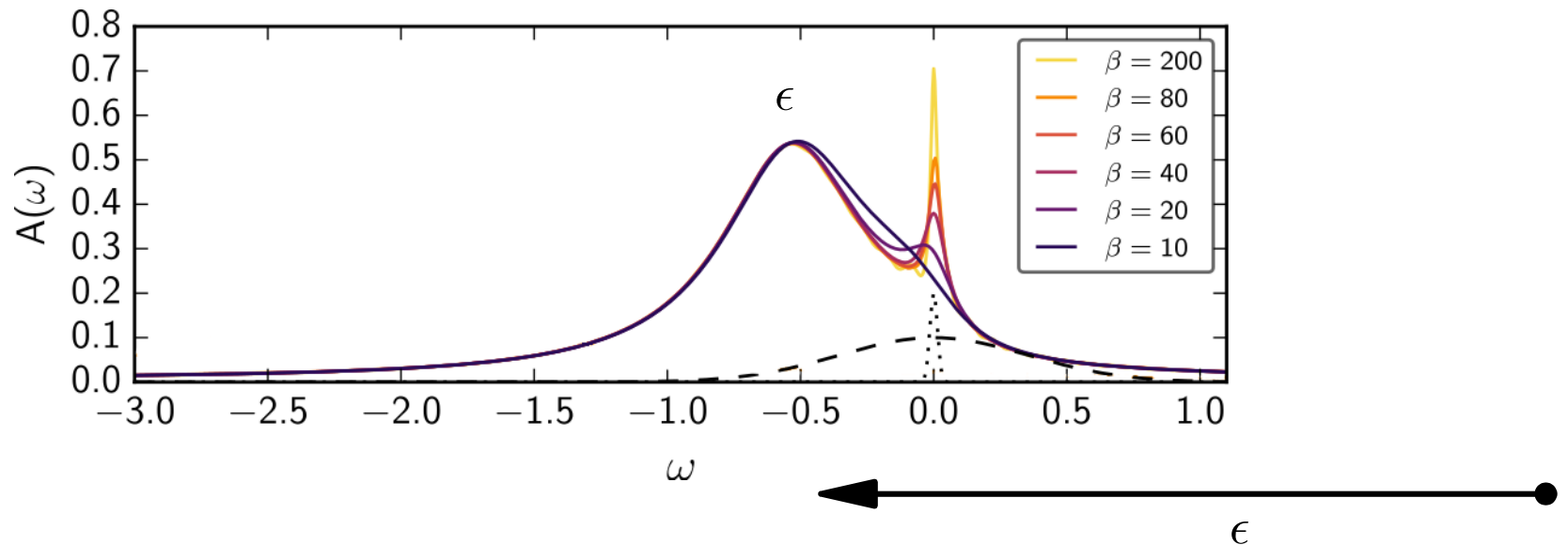


Buildup of the Kondo screening



Quench of the system towards the Kondo regime:
How quickly does the Kondo peak develop?

Buildup of the Kondo screening

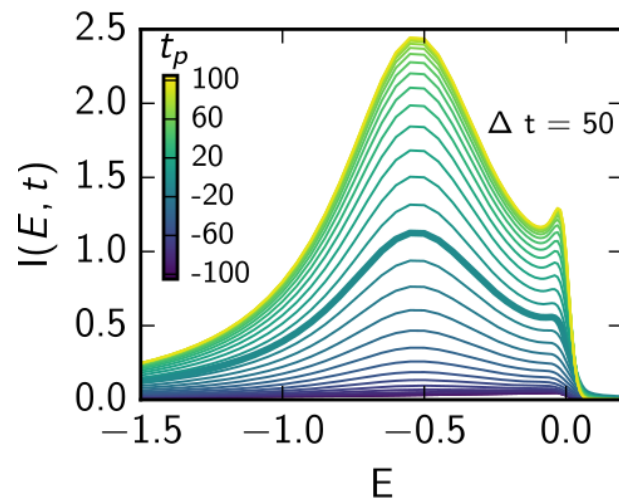
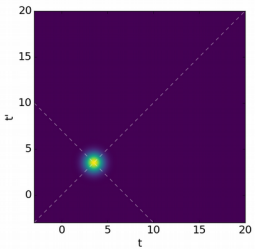
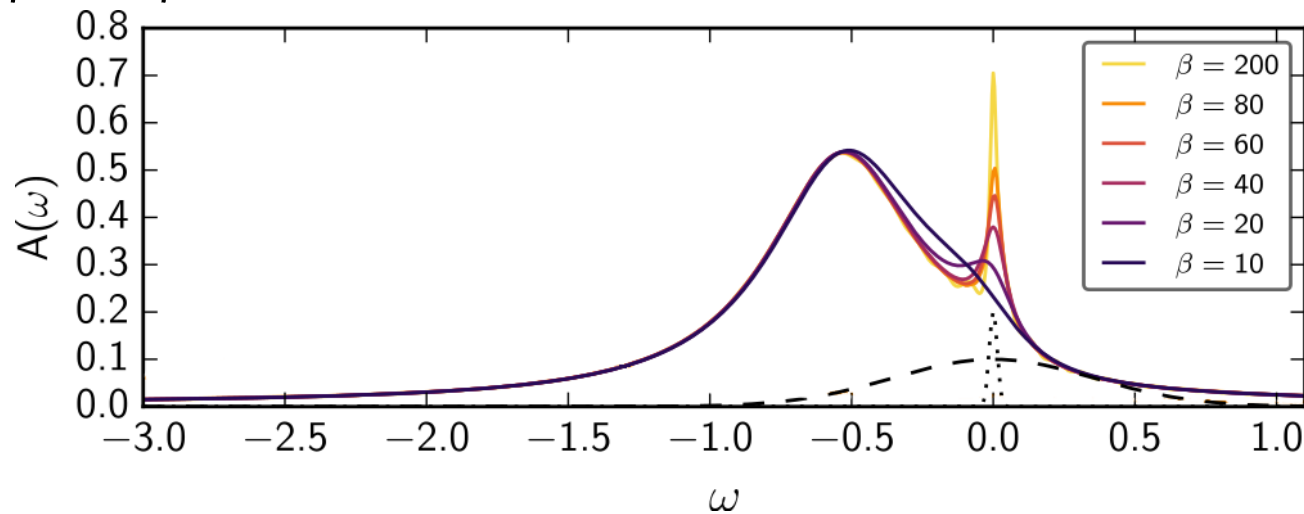


Time-resolved photoemission after quench of ϵ into the Kondo regime:

1. Standard single probe pulse
2. Two probe pulses

Buildup of the Kondo screening

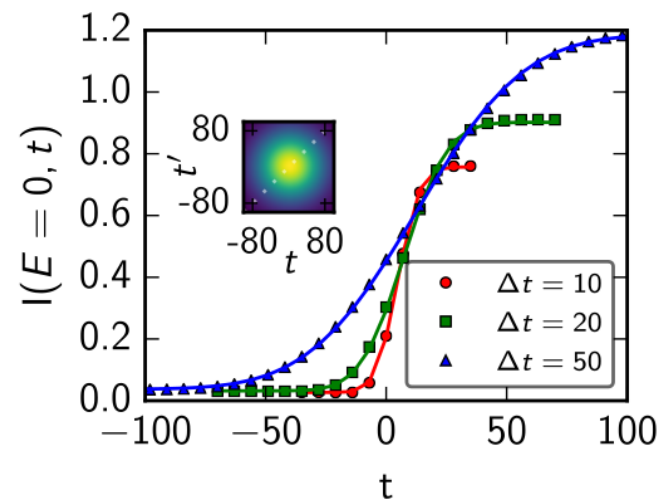
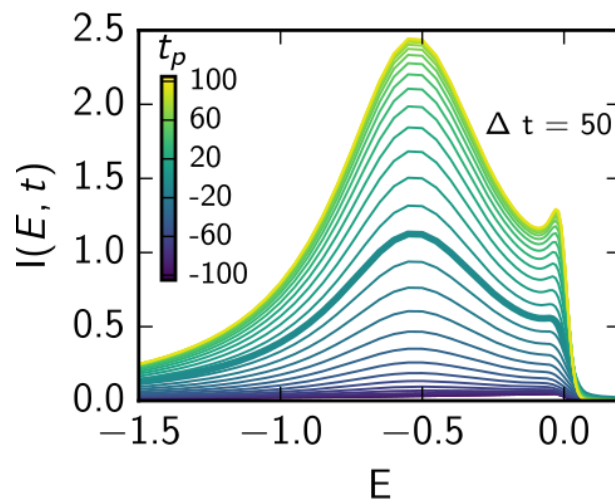
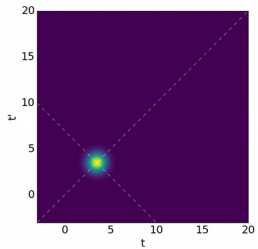
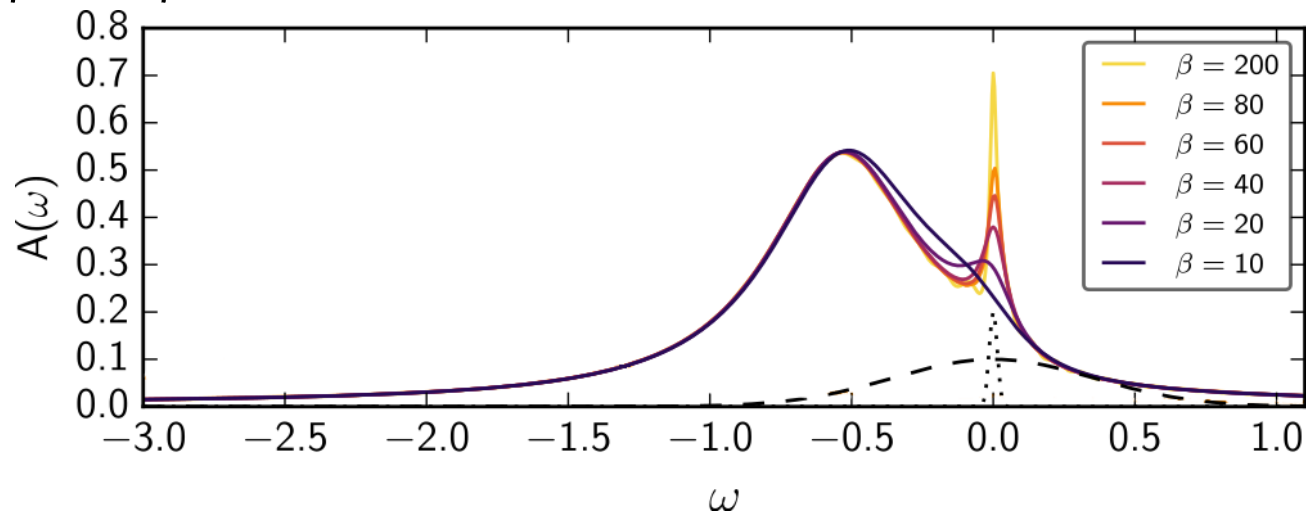
Standard single probe pulse



As first shown by Nordlander and Langreth (1999)

Buildup of the Kondo screening

Standard single probe pulse

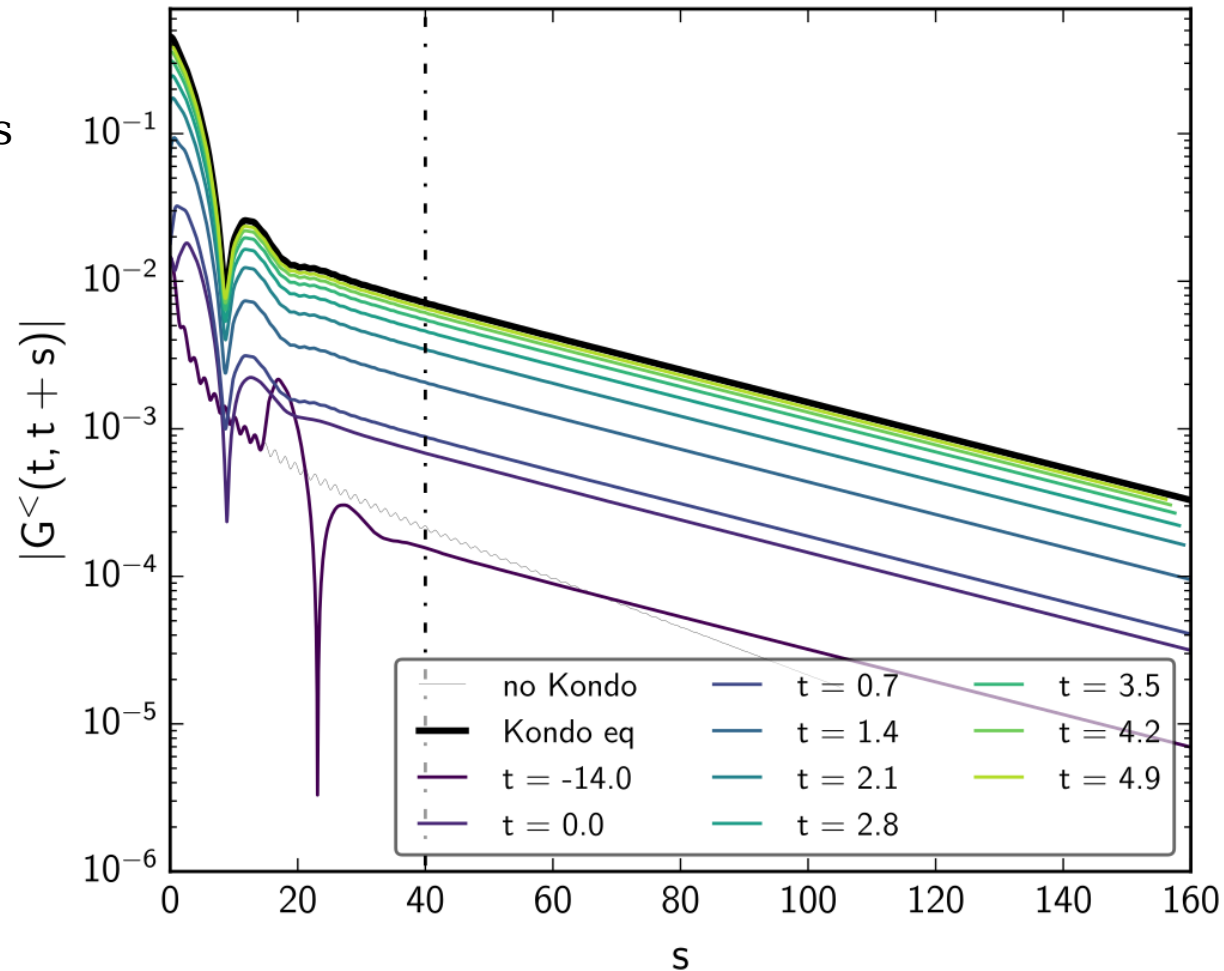


As first shown by Nordlander and Langreth (1999)

Buildup of the Kondo screening

True timescale of the screening
dictated by coupling to the electrons

→ faster than $\frac{\hbar}{\Delta E_{\text{Kondo}}}$

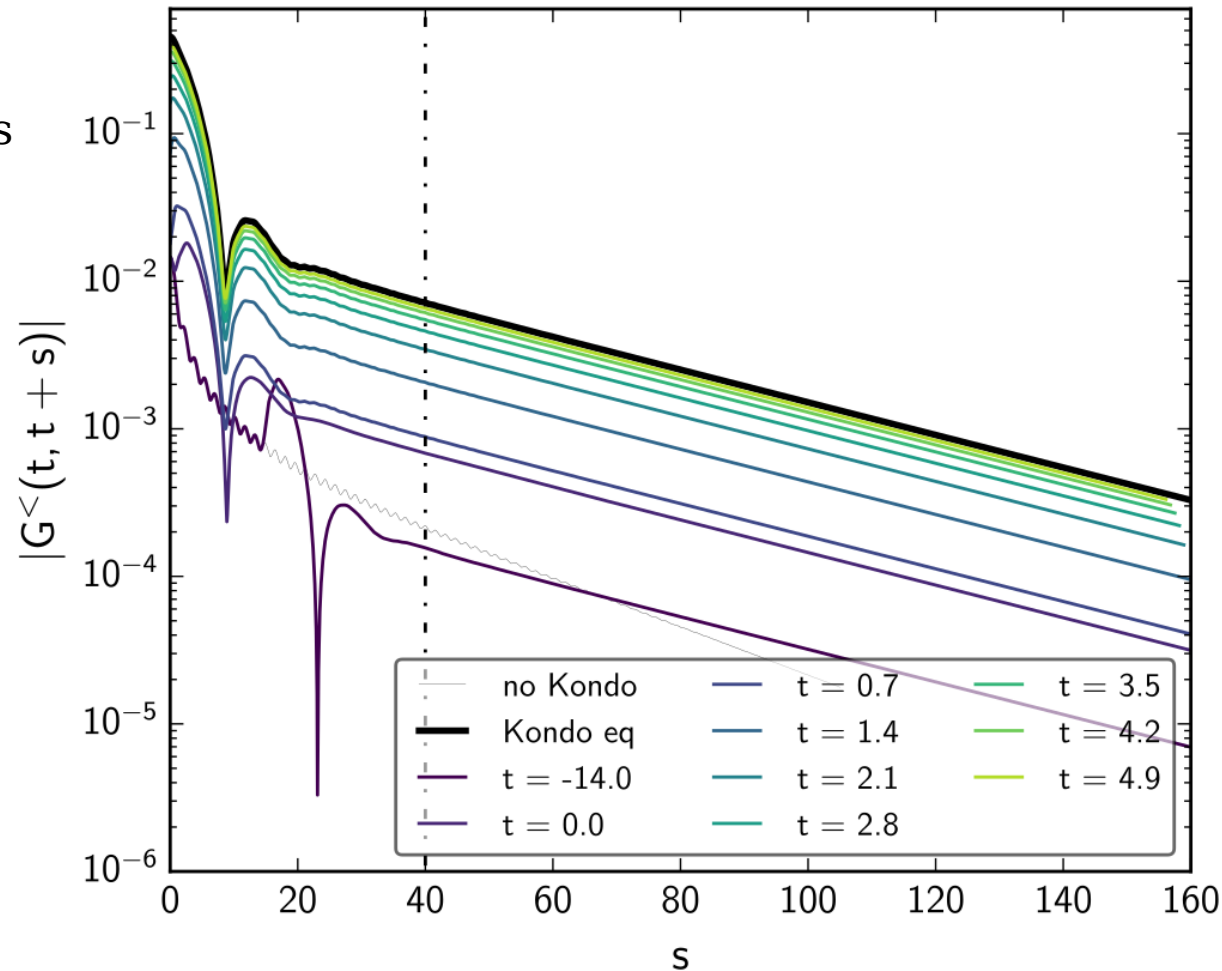


Buildup of the Kondo screening

True timescale of the screening dictated by coupling to the electrons

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Dynamics of the Kondo peak *beyond its spectral uncertainty limit*

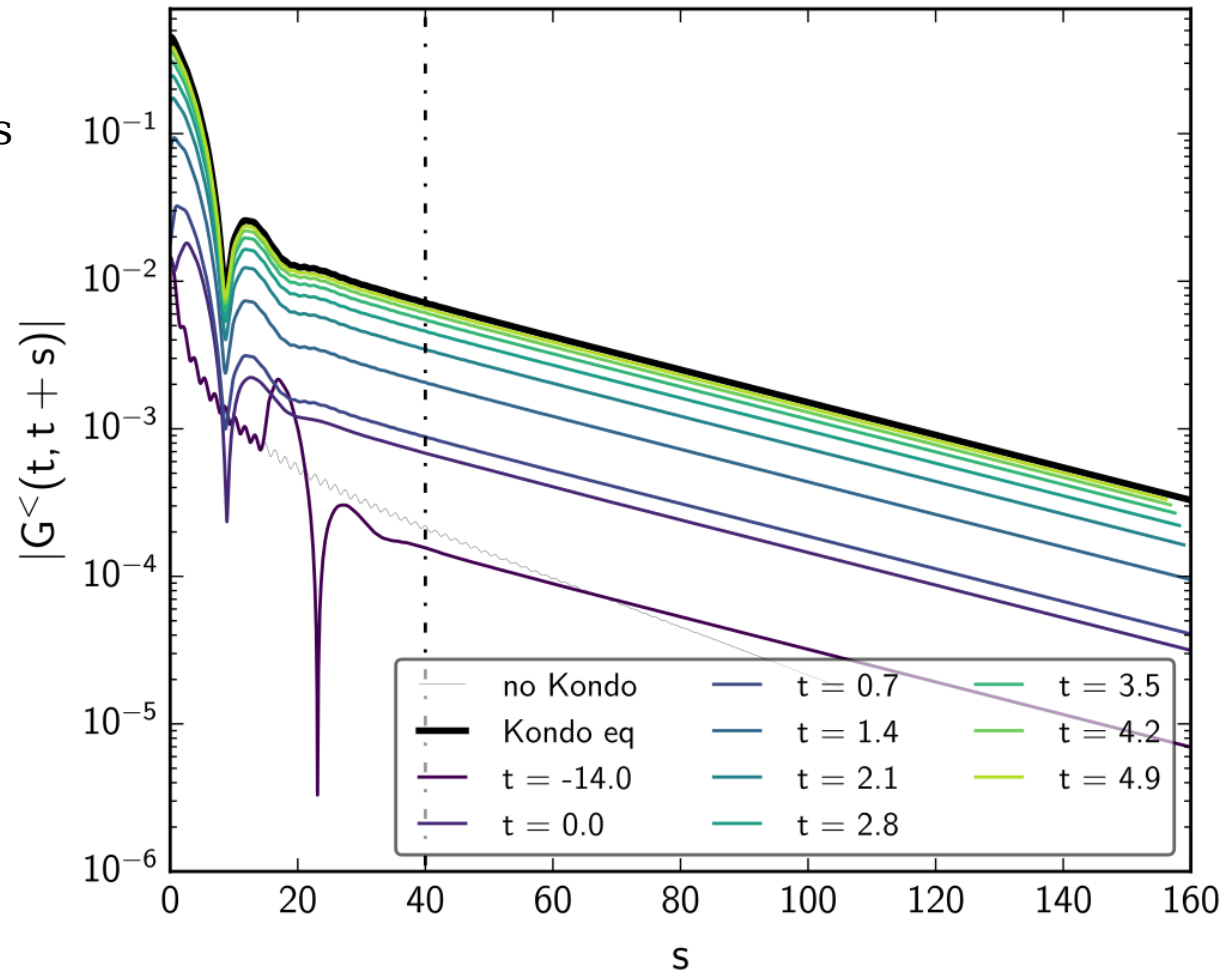


Buildup of the Kondo screening

True timescale of the screening
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Dynamics of the Kondo peak
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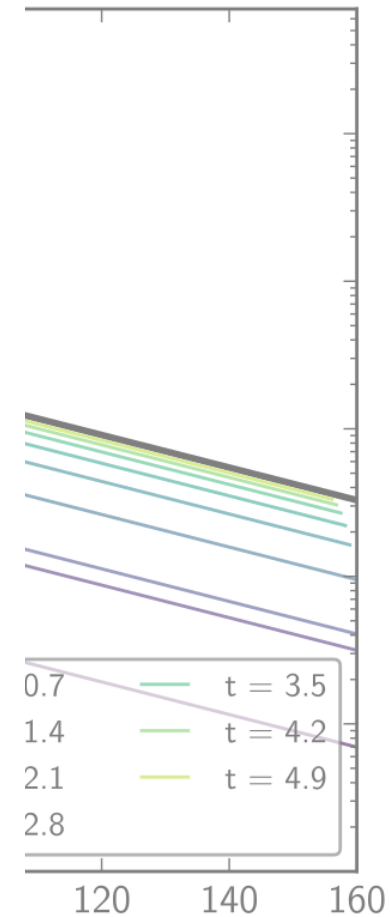
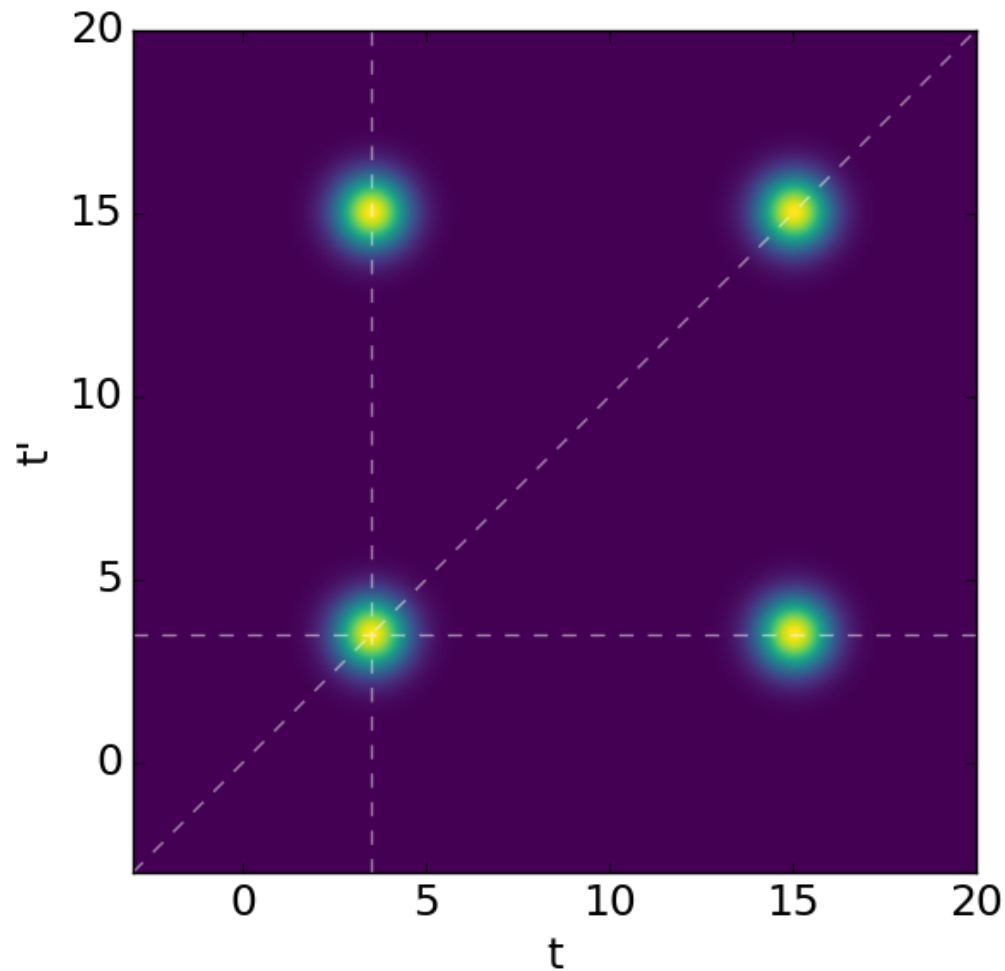


Buildup of the Kondo screening

True times
dictated by c

→ faster than

Dynamics
beyond its sp

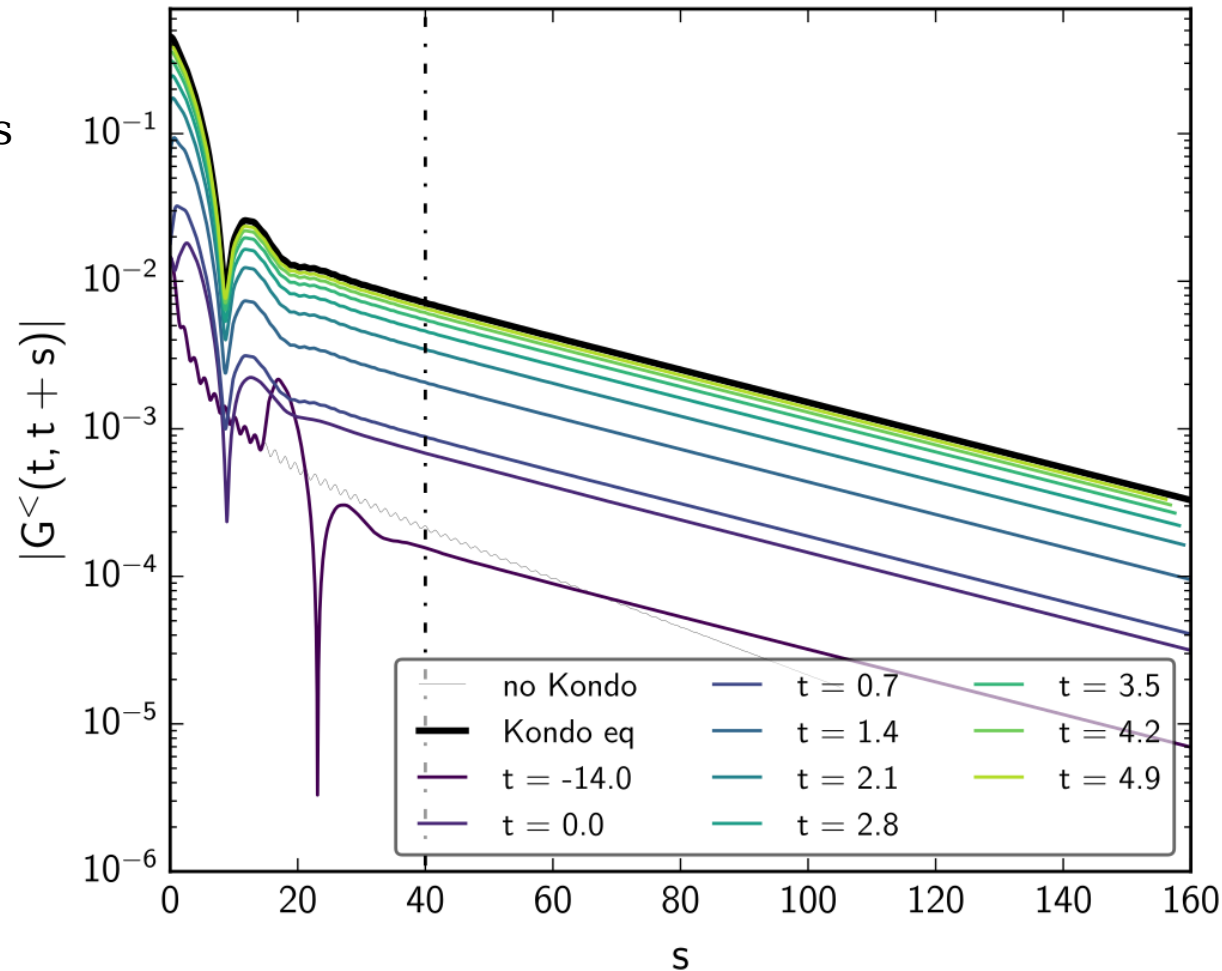


Buildup of the Kondo screening

True timescale of the screening
dictated by coupling to the electrons

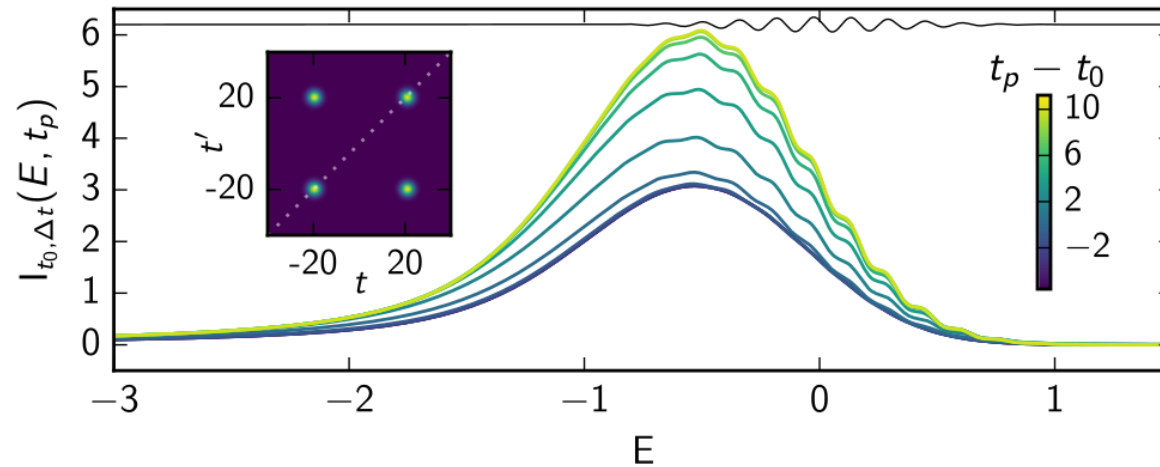
→ faster than $\frac{\hbar}{\Delta E_{\text{Kondo}}}$

Dynamics of the Kondo peak
beyond its spectral uncertainty limit



Buildup of the Kondo screening

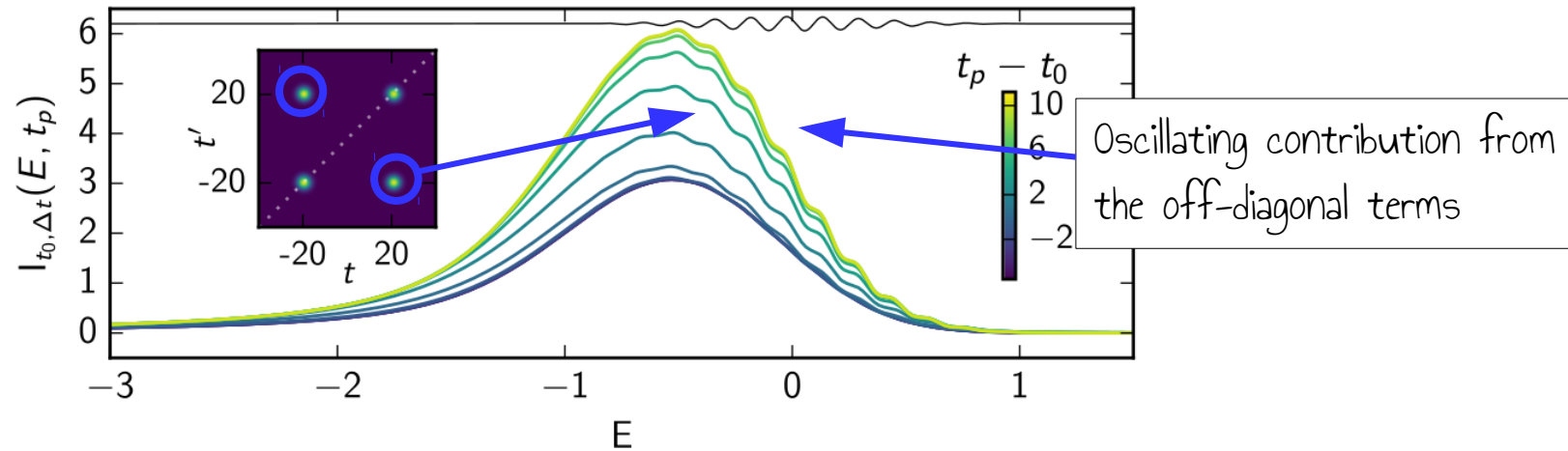
Two probe pulses



$$I(E, t_p) \propto \int dt dt' e^{-iE(t-t')} G^<(t_p + t, t_p - t') S(t, t')$$

Buildup of the Kondo screening

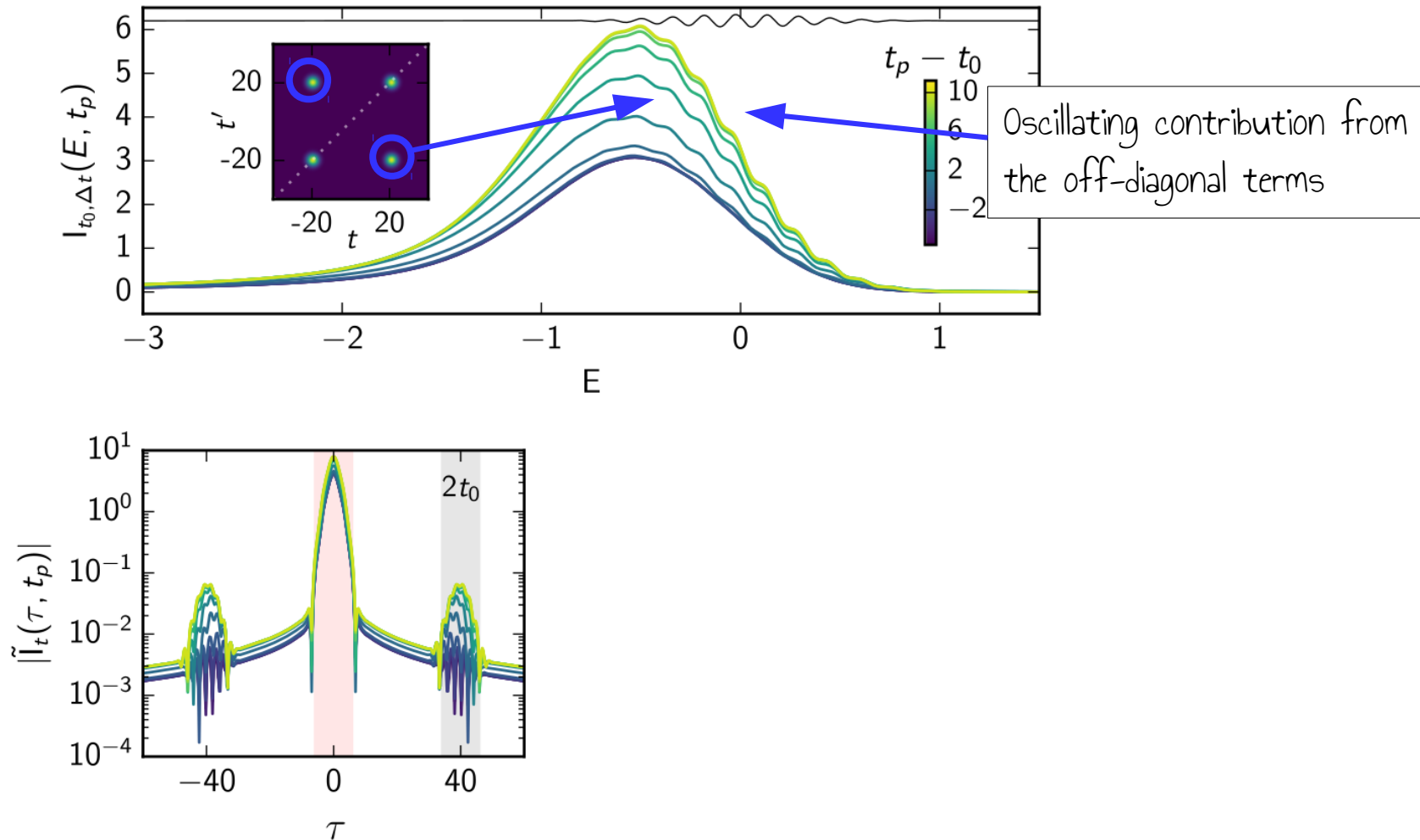
Two probe pulses



$$I(E, t_p) \propto \int dt dt' e^{-iE(t-t')} G^<(t_p + t, t_p - t') S(t, t')$$

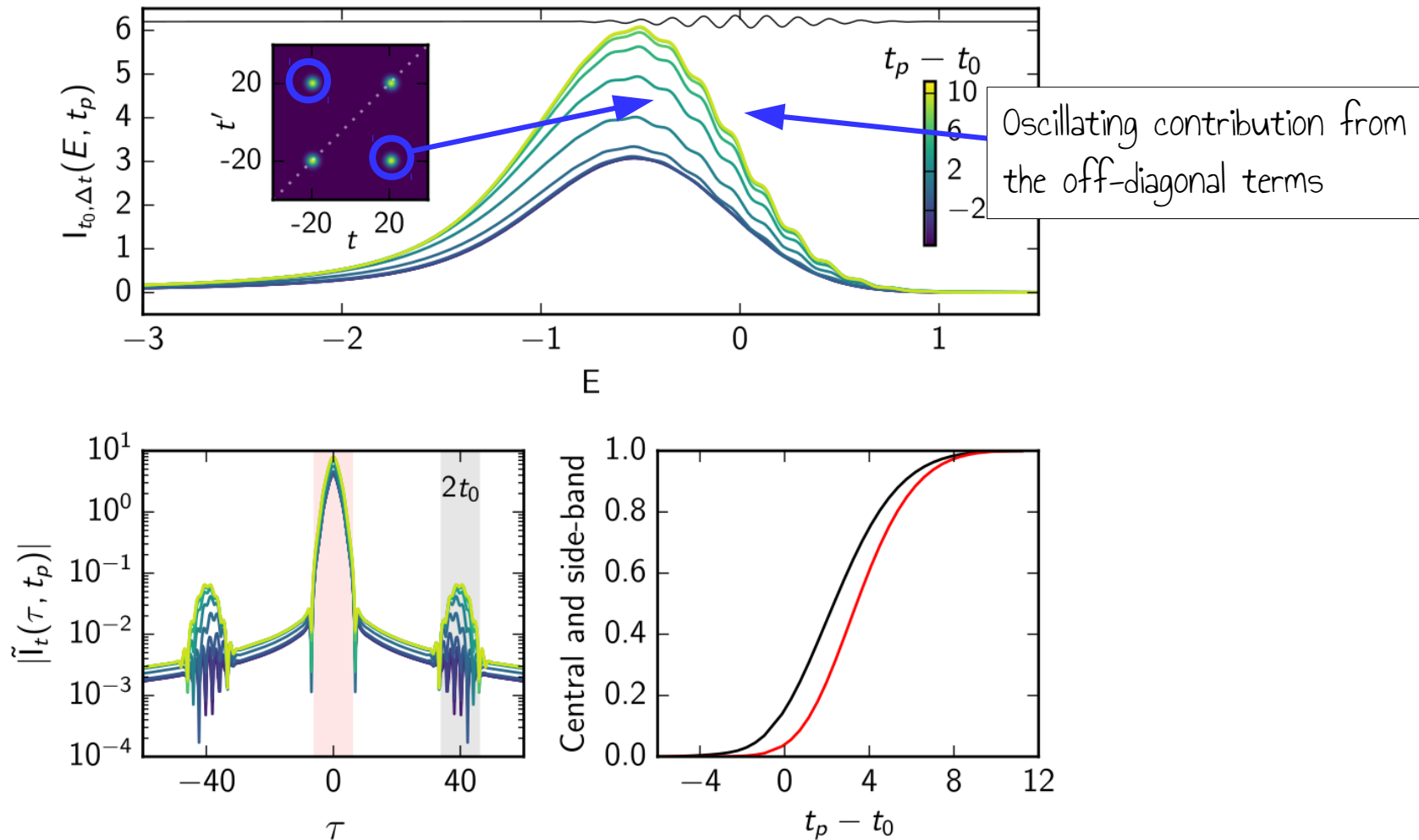
Buildup of the Kondo screening

Two probe pulses



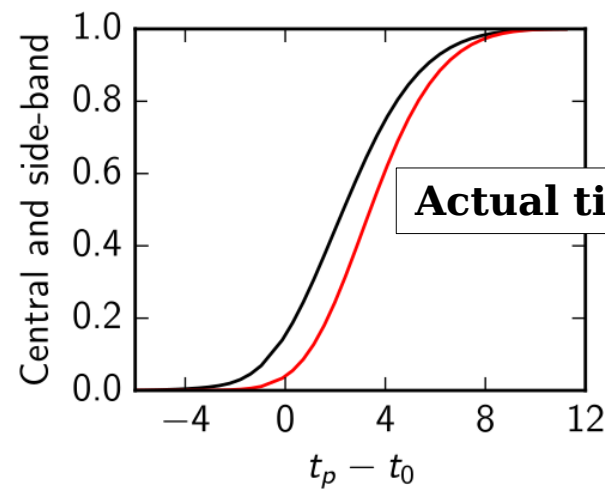
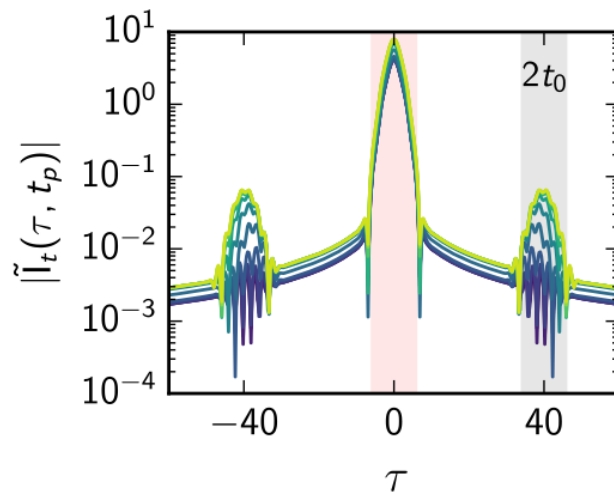
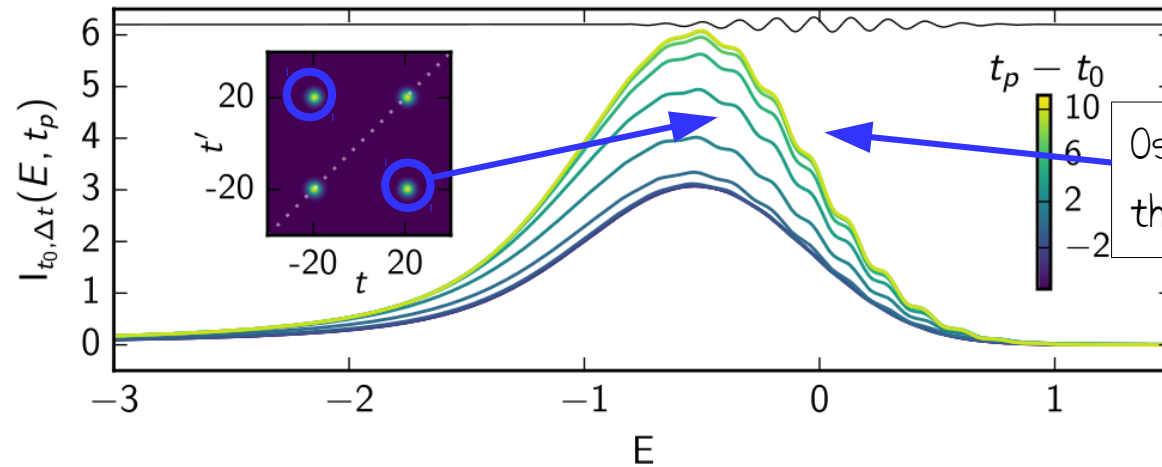
Buildup of the Kondo screening

Two probe pulses



Buildup of the Kondo screening

Two probe pulses



Conclusions

- **Dynamics beyond the spectral uncertainty limit**
common feature of strongly correlated materials
- **Energy-time uncertainty can be bypassed:** double pulse tomography – “two-dimensional” photoemission

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Thank you