

# Spectrally-resolved femtosecond reflectivity relaxation dynamics in undoped SDW 122-structure iron based pnictides

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A description of this work can be found in:

PRB **81**, 224504 (2010); PRB **82**, 012505 (2010); PRB **86**, 024519 (2012);  
PRB **89**, 165131 (2014)

## Acknowledgements

optical:

A. Pogrebna, L. Stojchevska, N. Vujičić,

V. Kabanov, D. Mihailovic, JSI, Ljubljana

samples:

122:

I. R. Fisher, J.-H. Chu, Stanford University, Stanford, USA, (Ba-122)

Z. A. Xu, X. Lin, G. H. Cao, Zhejiang University, Hangzhou, People's Republic of China, (Sr-122, Eu-122)

1111:

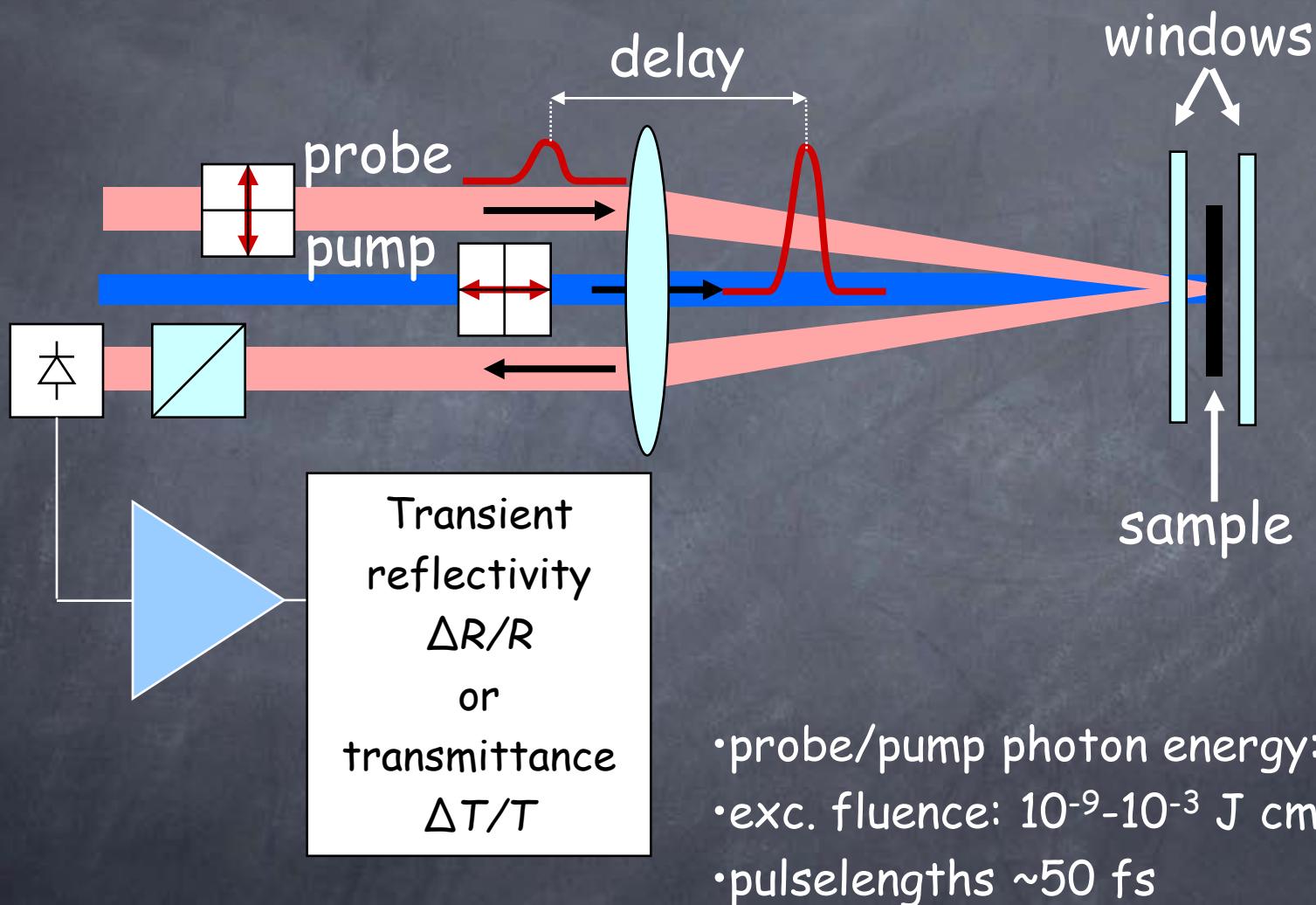
J. Karpinski, N.D. Zhigadlo,

Z. Bukowski, S. Katrych, Laboratory for Solid State Physics, ETH Zürich, Switzerland and LPMC, EPFL, Lausanne, Switzerland

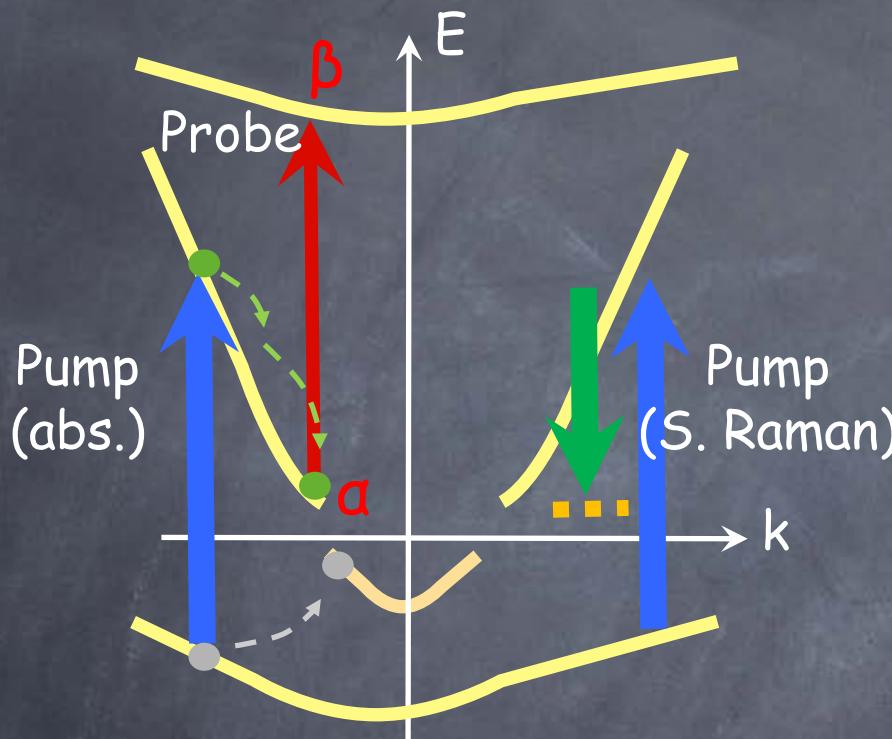
# Outline

- Introduction to all-optical transient spectroscopy
- Quasiparticle relaxation bottleneck in systems with small gap: the Rothwarf-Taylor bottleneck
- Dynamics of the CDW state. Is there a Rothwarf-Taylor bottleneck at all?
- Dynamics of the SDW state in iron based pnictides
- Ultrafast SDW state destruction and recovery
- Conclusions

# Time resolved optical spectroscopy setup



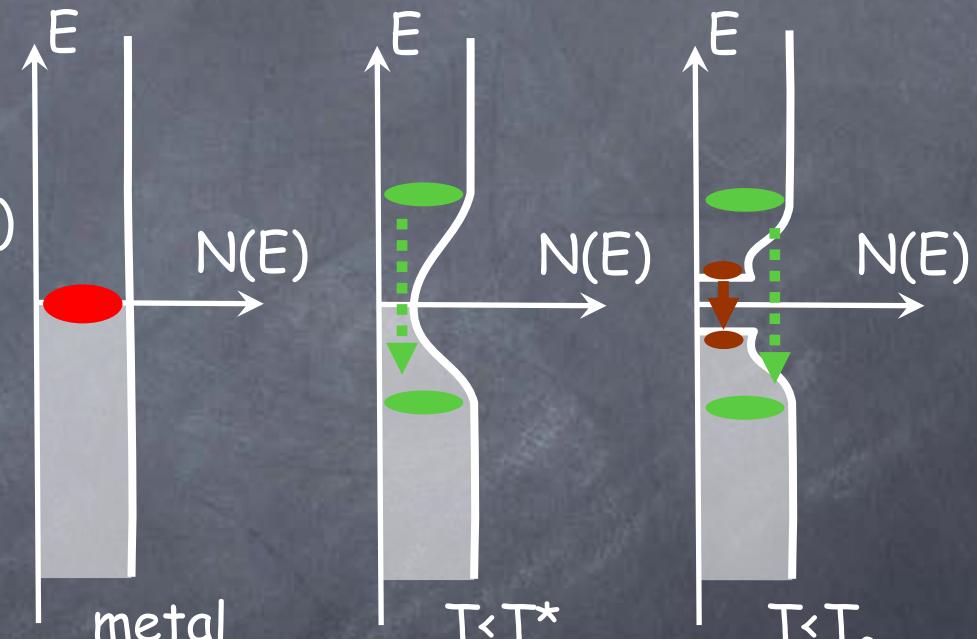
# Transient reflectivity (transmittance)



The rigid band approximation:

$$\Delta R_{\alpha,\beta} \propto \int d^3k [ |M_{\alpha,\beta}(\mathbf{k})|^2 \Delta f_\alpha(\mathbf{k}) \\ \times g(\epsilon_\beta(\mathbf{k}) - \epsilon_\alpha(\mathbf{k}) - \hbar\omega_{\text{probe}}) ].$$

The transient reflectivity probes the photoexcited QP density.



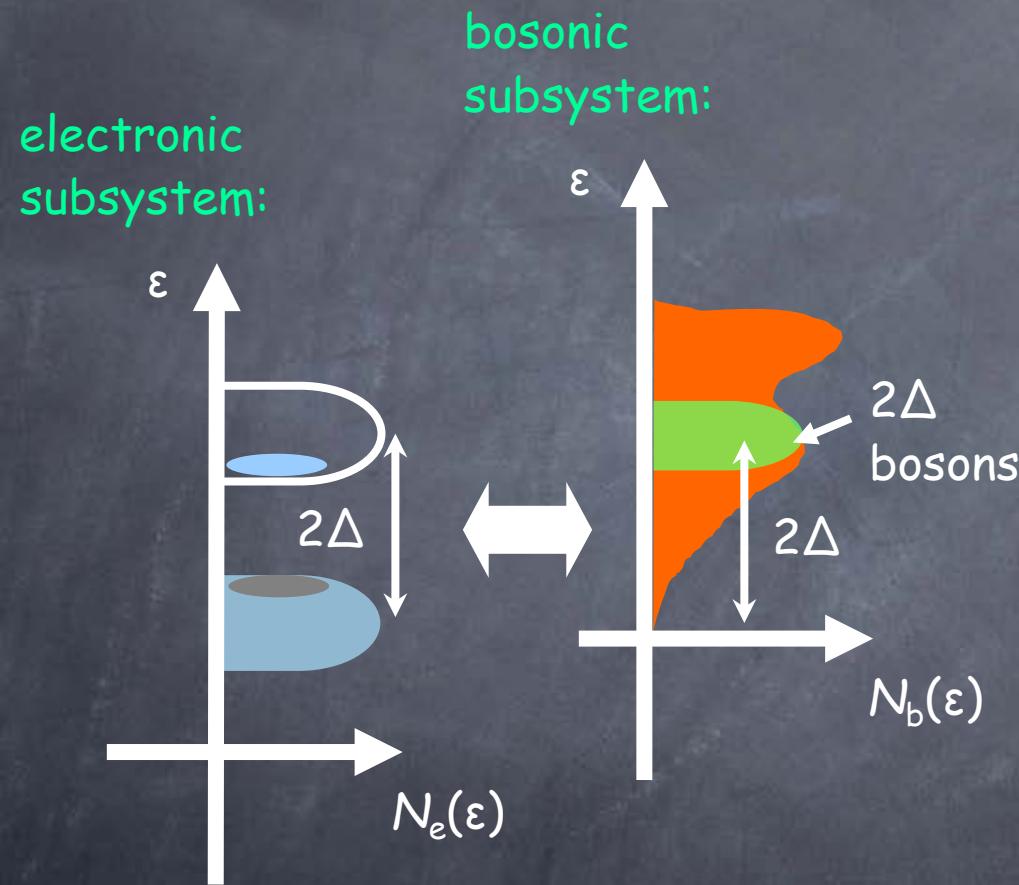
e-ph coupling

$\Delta_{\text{PG}}$

$\Delta_{\text{PG}}, \Delta_{\text{SC}}, \Delta_{\text{CDW}}$

# Bottleneck in systems with small gap

(weak excitation) [Rothwarf-Taylor 1967, Kabanov 1999]



**Energy conservation:**

$$E_L = [c_e(T_{\text{bath}}) + c_{2\Delta b}(T_{\text{bath}})] \delta T_{el}$$

$$n_{pe}(T) = dn/dT \quad \delta T_{el}$$

**BCS  $T$ -dependent gap:**

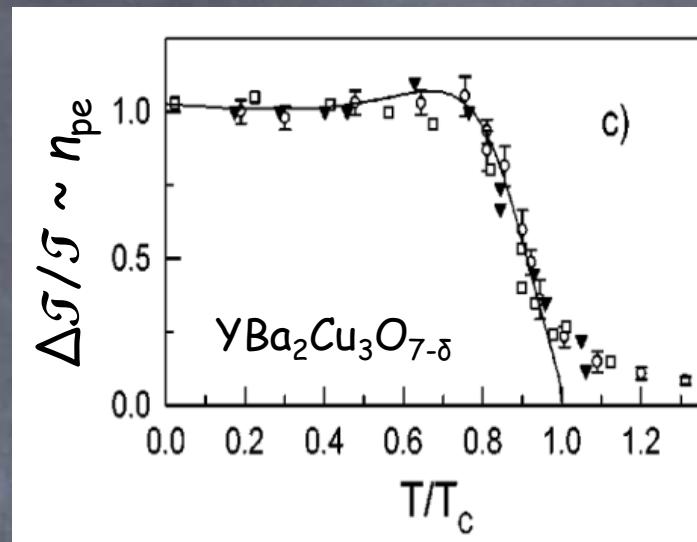
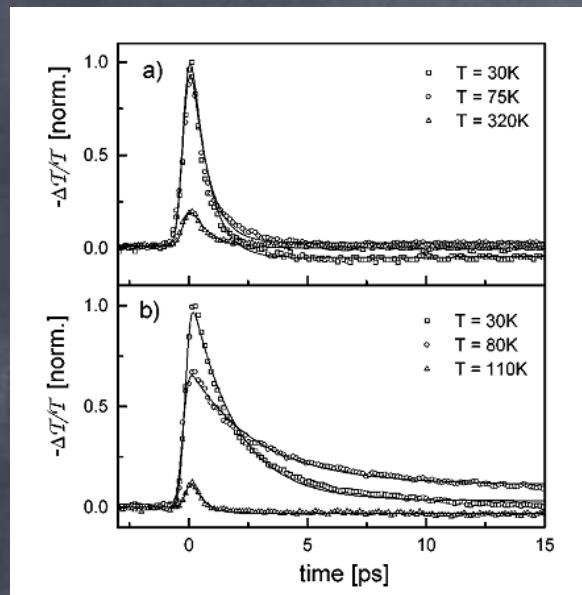
$$n_{pe} = \frac{\mathcal{E}_I / (\Delta(T) + k_B T/2)}{1 + \frac{2\nu}{N(0)\hbar\Omega_c} \sqrt{\frac{2k_B T}{\pi\Delta(T)}} \exp[-\Delta(T)/k_B T]}.$$

V.V. Kabanov et al., PRB 59, 1497 (1999)

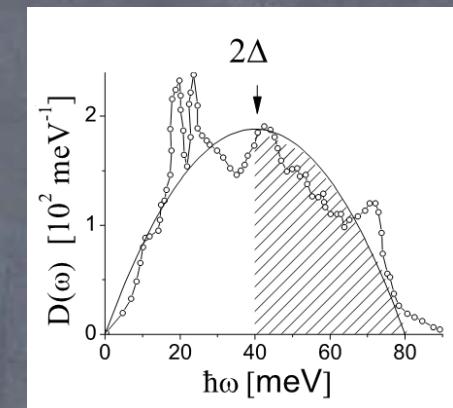
$N(0)\hbar\Omega_c$  no. of involved electronic degrees of freedom

$\nu$  no. of involved boson degrees of freedom

# Bottleneck in superconductors



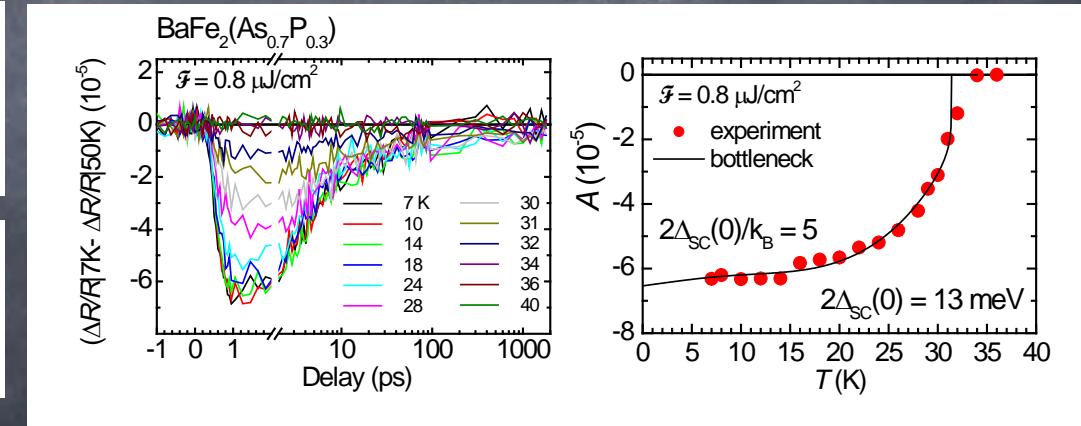
V.V. Kabanov et al., PRB 59, 1497 (1999)



B. Renker et al.,  
Physica B 165-166, 1237 (1990)

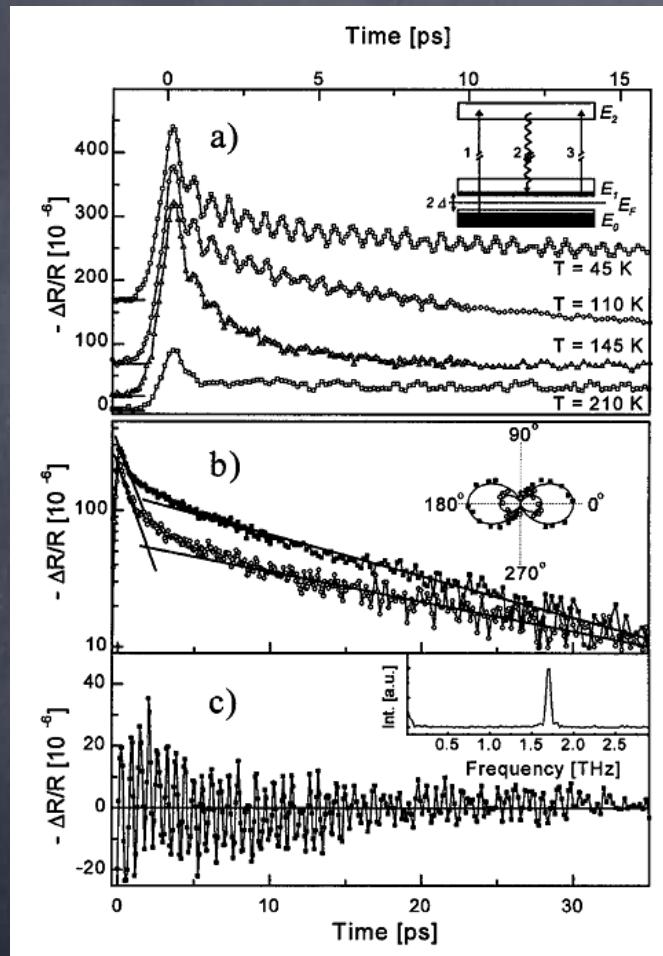
$$\Delta R_{\alpha,\beta} \propto \int d^3k [ |M_{\alpha,\beta}(\mathbf{k})|^2 \Delta f_\alpha(\mathbf{k}) \times g(\epsilon_\beta(\mathbf{k}) - \epsilon_\alpha(\mathbf{k}) - \hbar\omega_{\text{probe}})].$$

$$n_{pe} = \frac{\mathcal{E}_I / (\Delta(T) + k_B T/2)}{1 + \frac{2\nu}{N(0)\hbar\Omega_c} \sqrt{\frac{2k_B T}{\pi\Delta(T)}} \exp[-\Delta(T)/k_B T]}.$$

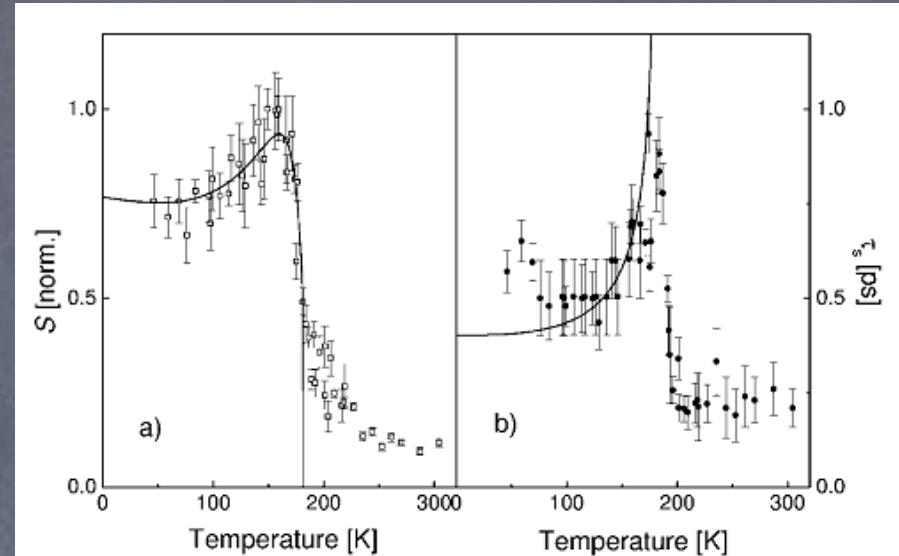


# Application of the bottleneck model to a CDW

$K_{0.3}MoO_3$



J. Demsar et al., PRL 83, 800 (1999)

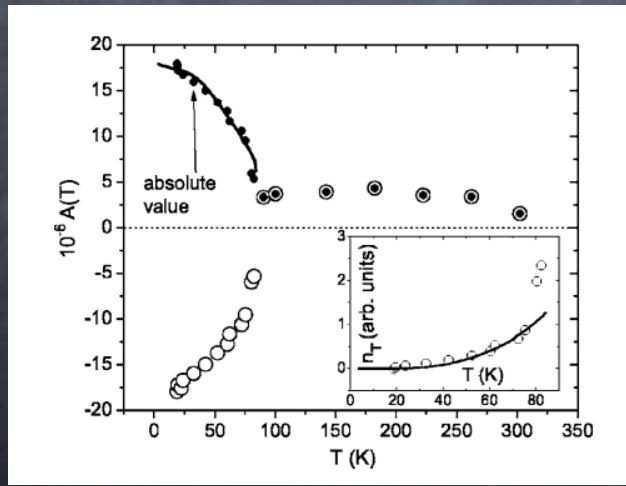
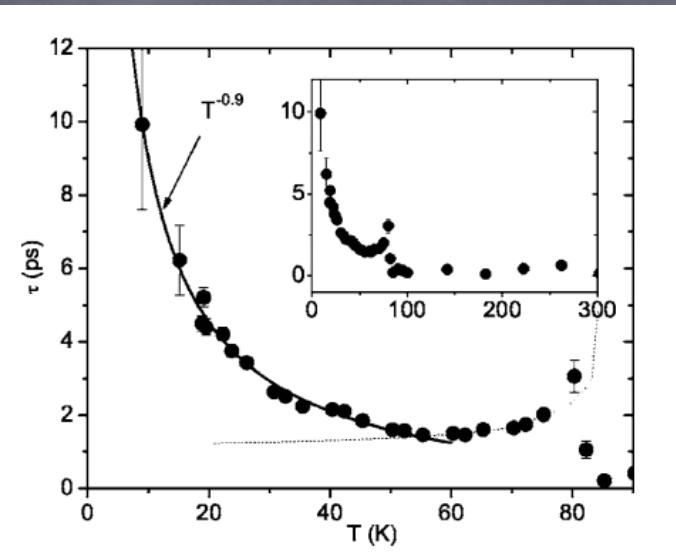
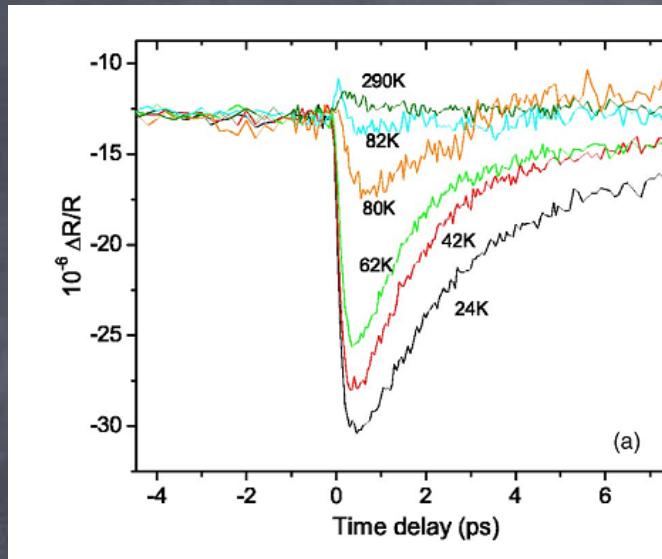


$$n_{pe} = \frac{\mathcal{E}_I / (\Delta(T) + k_B T/2)}{1 + \frac{2\nu}{N(0)\hbar\Omega_c} \sqrt{\frac{2k_B T}{\pi\Delta(T)}} \exp[-\Delta(T)/k_B T]}.$$

$2\Delta(0) \sim 150 \text{ meV} > \text{max. phonon energy}$

# Application of the bottleneck model to a SDW

$\text{UNiGa}_5 \quad T_N = 85 \text{ K}$

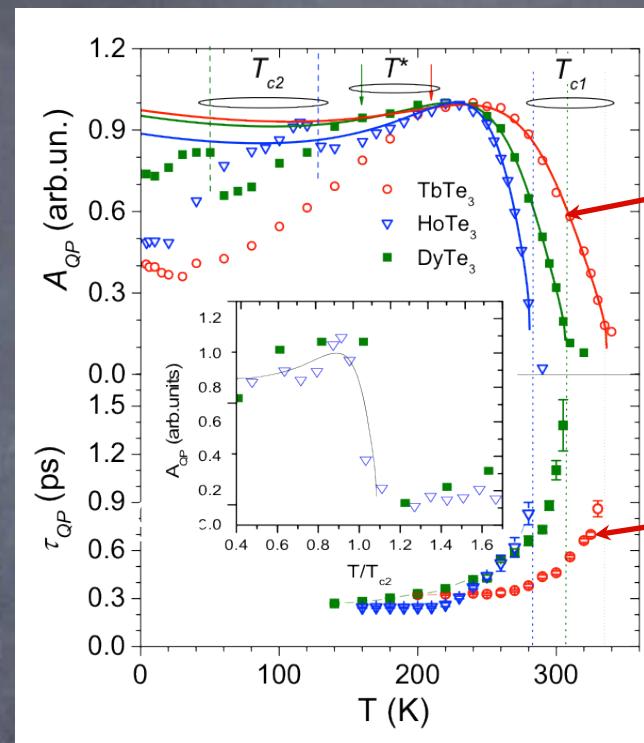
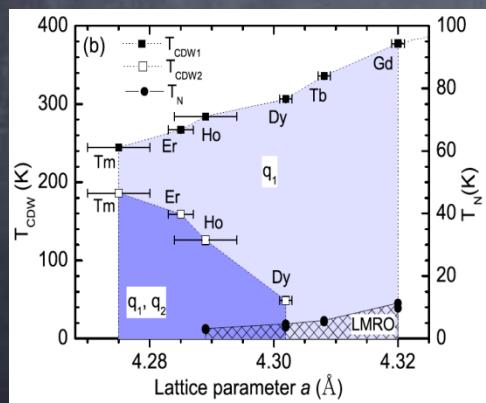
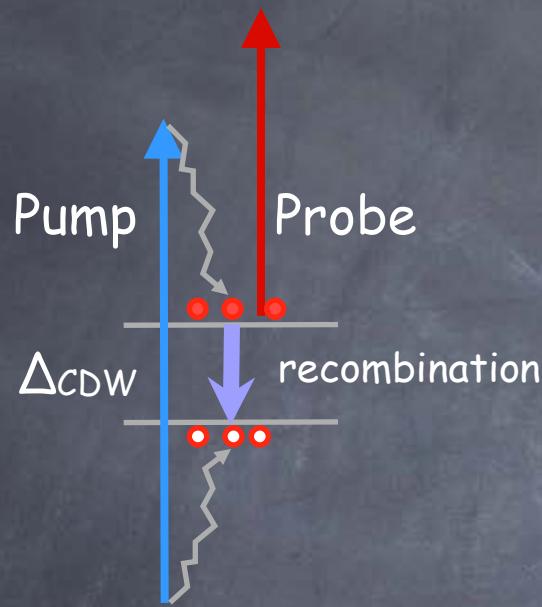


$$|A(T)| \propto \frac{\epsilon_F [\Delta(T) + k_B T / 2]}{1 + B \sqrt{\frac{2k_B T}{\pi \Delta(T)}} \exp\left(\frac{-\Delta(T)}{k_B T}\right)}$$

$$\Delta_0 = 1.7k_B T_N$$

E. E. M. Chia et al., PRB 74, 140409 (2006)

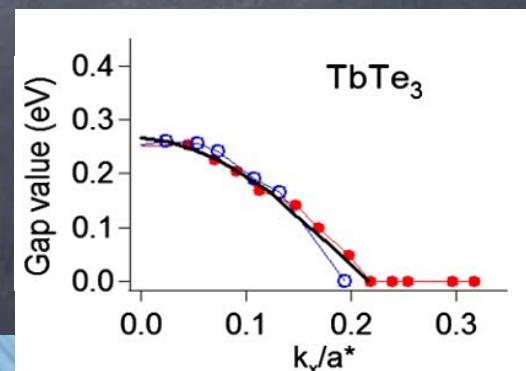
# Bottleneck model in CDW state of tri-tellurides



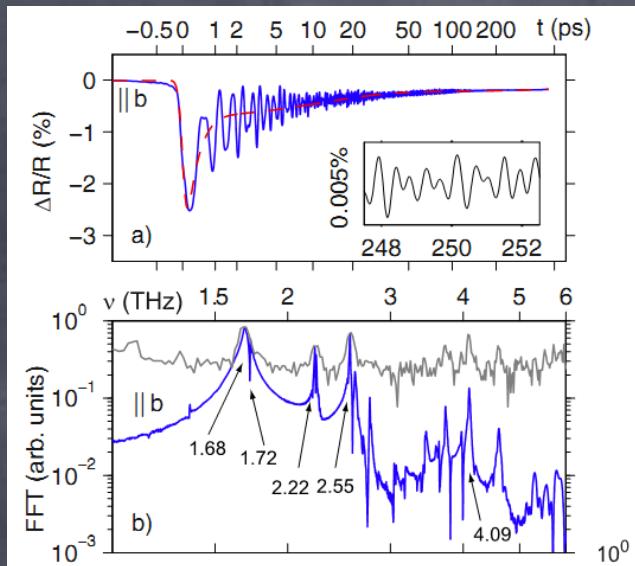
The reflectivity amplitude  
(BCS-like T-dependence of  
the CDW gap)

The relaxation rate  
 $1/\tau \propto \Delta(T)$

**TbTe<sub>3</sub>:**  
 $\Delta(0) \sim 125$  meV



# Quasiparticle vs collective mode dynamics in CDW?

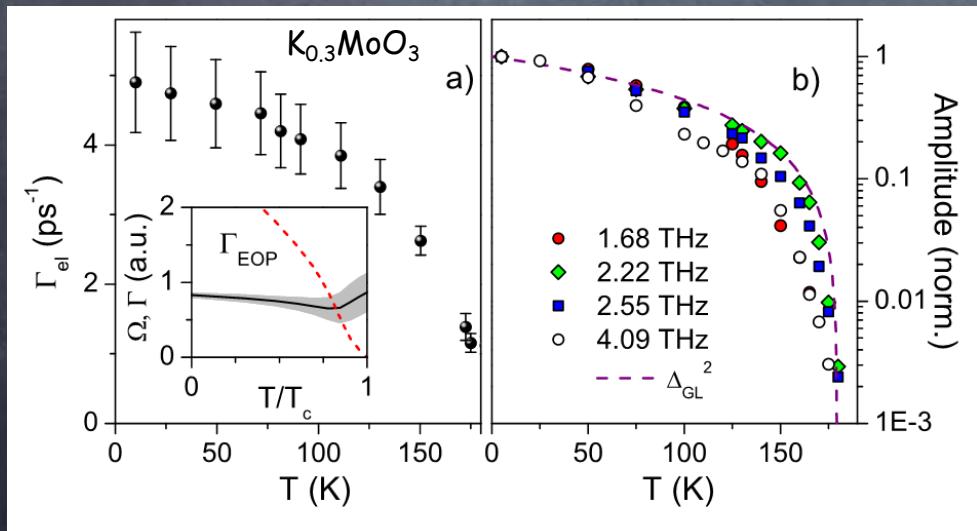


Overdamped electronic order parameter:  $x = \psi(t) - \Psi_{eq}$

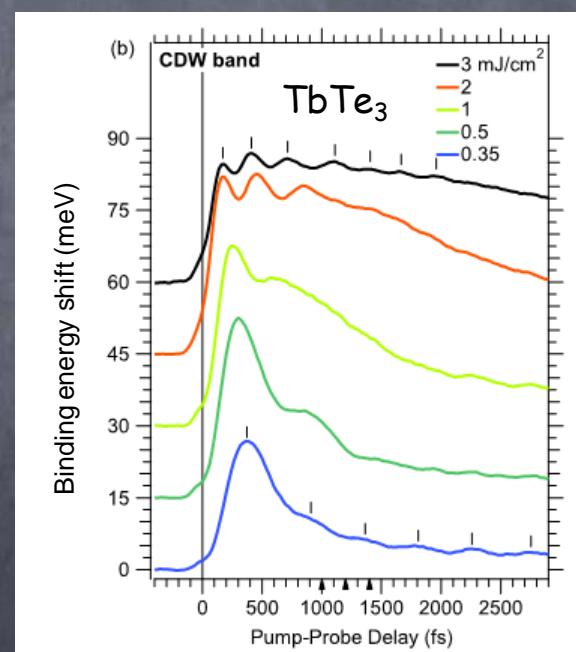
$$\dot{x}_1 = -2\kappa\alpha\left(T_c - T + \frac{m^2}{2\alpha\Omega_0^2}\right)x_1 - \kappa m y_1$$

$$\ddot{y}_1 = -\Omega_0^2 y_1 - mx_1,$$

Phonon(s)



Schaefer et al., PRL 105, 066402 (2010)

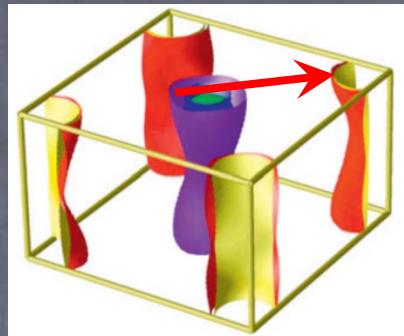
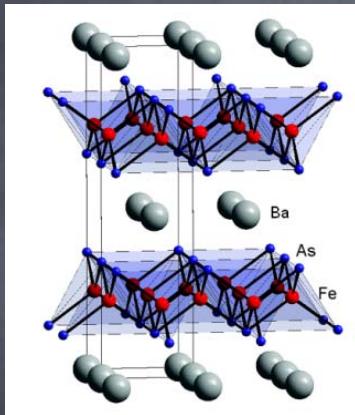


F. Schmitt et al., New Journal of Physics 13, 063022 (2011)

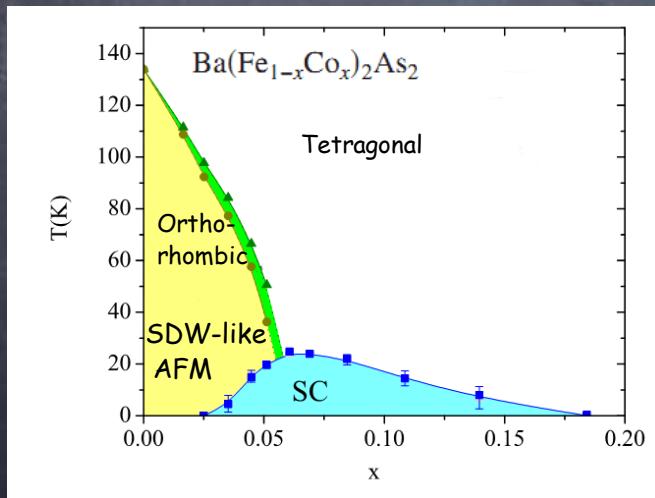
# Fe based pnictides

Ba-122:

- $\text{Ba}(\text{Fe},\text{Co})_2\text{As}_2$ ,  $T_{c,\text{max}} \approx 24\text{K}$



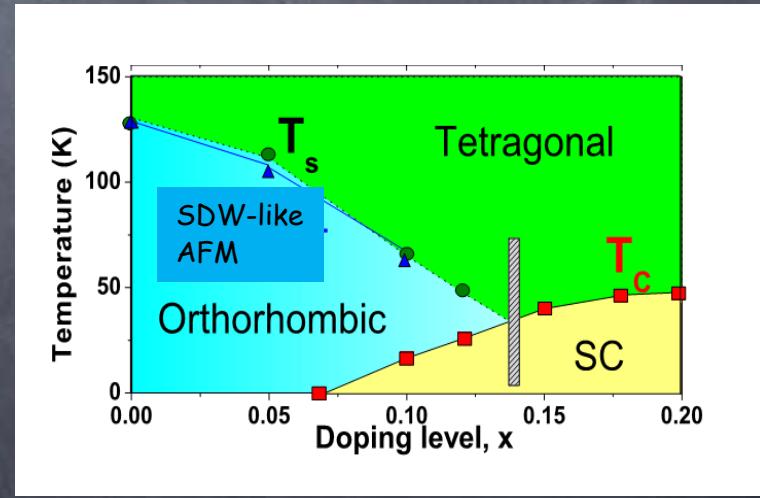
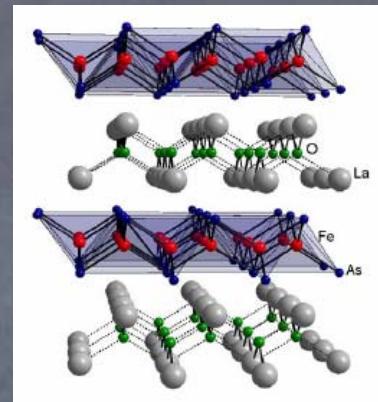
I.I. Mazin et al.  
Physica C 469, 614-627 (2009)



J.-H. Chu et al., Phys. Rev. B 79, 014506 (2009)

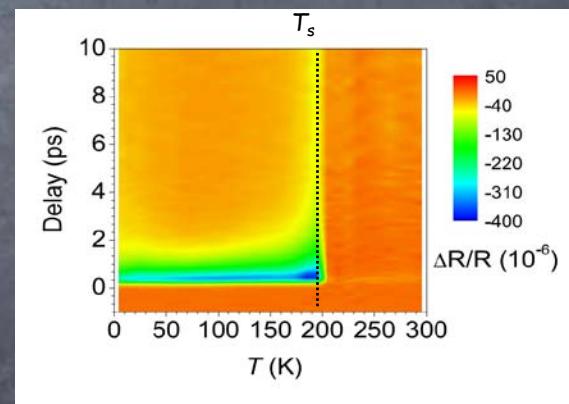
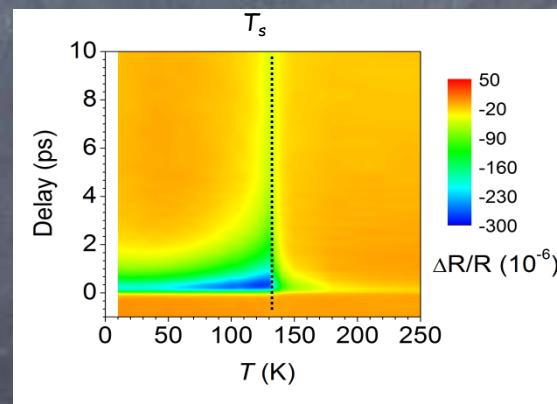
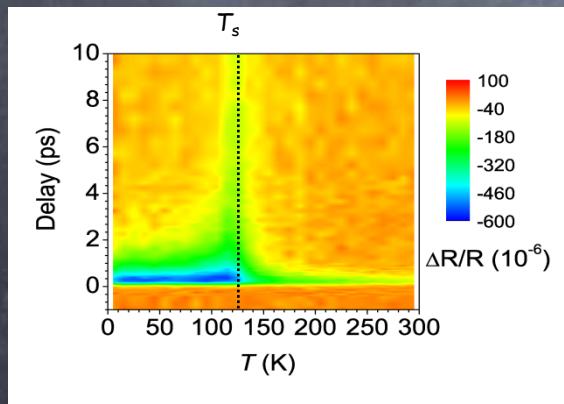
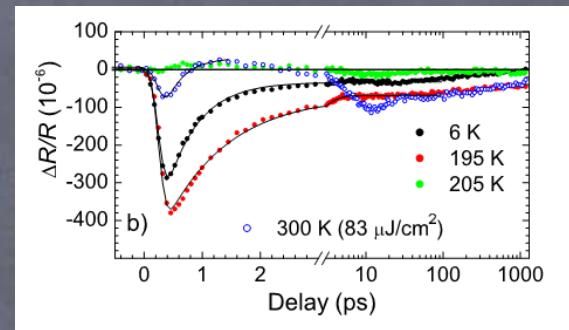
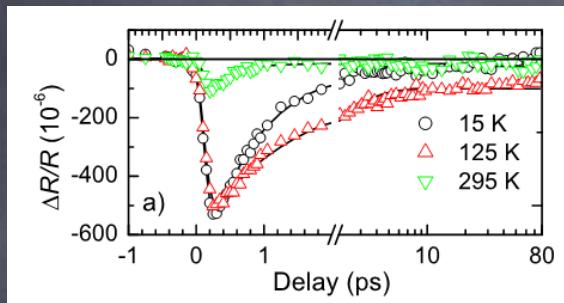
Sm-1111:

- $\text{SmFeAs(O,F)}$ ,  $T_{c,\text{max}} \approx 55\text{K}$
- $\text{Sm}(\text{Fe},\text{Co})\text{AsO}$ ,  $T_{c,\text{max}} \approx 17\text{K}$



Mrgadonna et al., Phys. Rev. B 79, 014503 (2009)

# Comparison of $\Delta R/R$ transients in undoped (SDW) compounds (1.55 eV probe)



$\text{SmFeAsO}_{1-\delta}$

$T_N < T_s \approx 125\text{ K}$

$\text{BaFe}_2\text{As}_2$

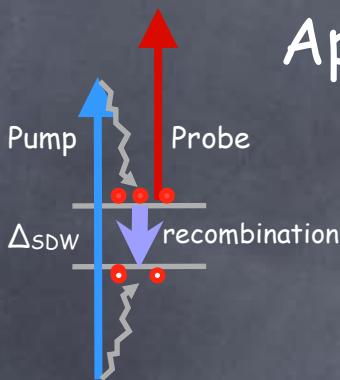
$T_N = T_s \approx 135\text{ K}$

$\text{SrFe}_2\text{As}_2$

$T_N = T_s \approx 200\text{ K}$

Stojchevska et al., PRB **82**, 012505 (2010)

# Application of the bottleneck model to SDW

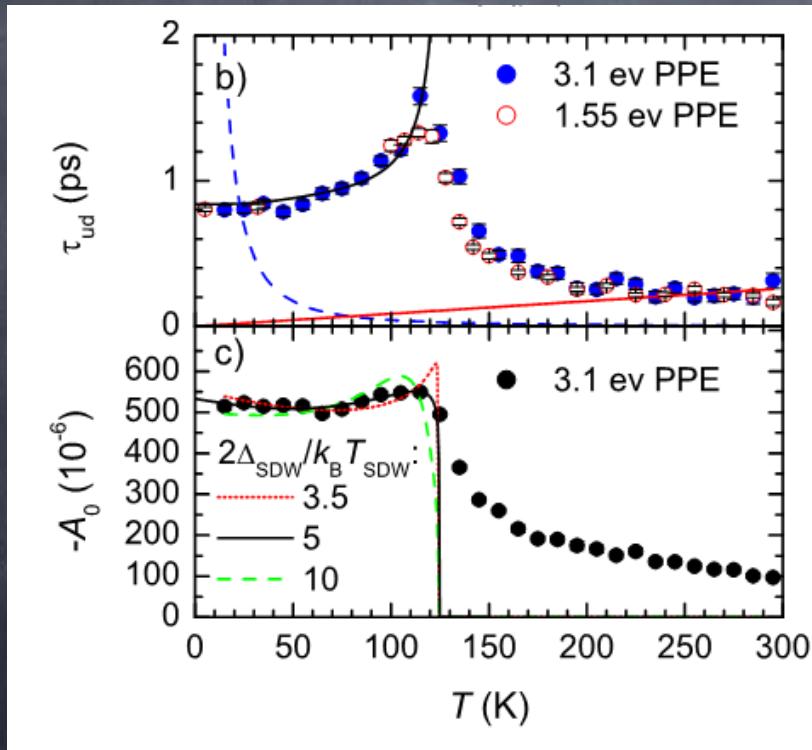


BCS  $T$ -dependent gap:

$$n_{pe} = \frac{\mathcal{E}_I / (\Delta(T) + k_B T/2)}{1 + \frac{2\nu}{N(0)\hbar\Omega_c} \sqrt{\frac{2k_B T}{\pi\Delta(T)}} \exp[-\Delta(T)/k_B T]}.$$

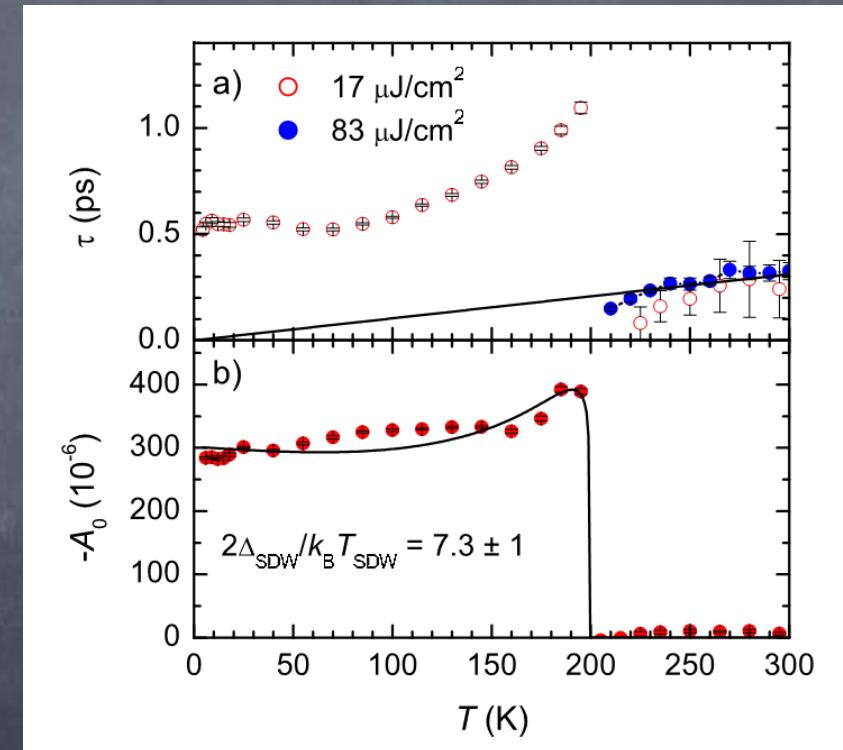
V.V. Kabanov et al., PRB 59, 1497 (1999)

$SmFeAsO_{1-\delta}$



Mertelj et al., PRB 81, 224504 (2010)

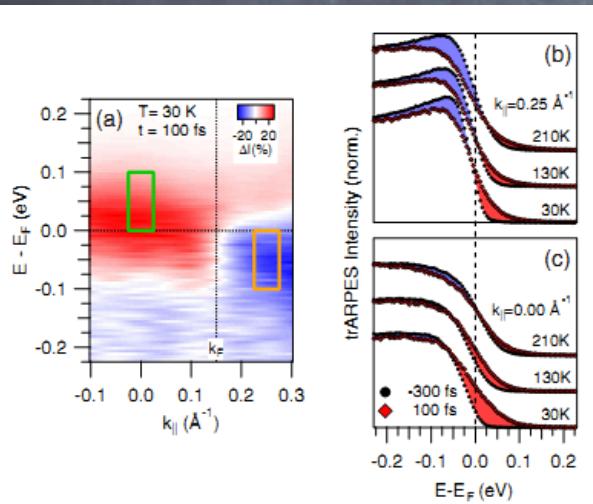
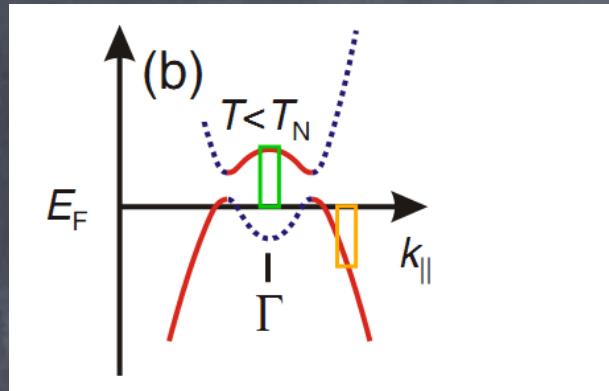
$SrFe_2As_2$



Stojchevska et al., PRB 82, 012505 (2010)

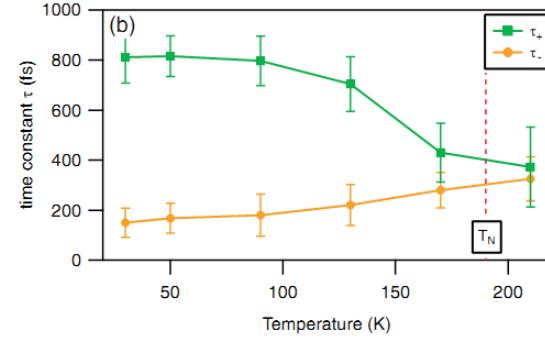
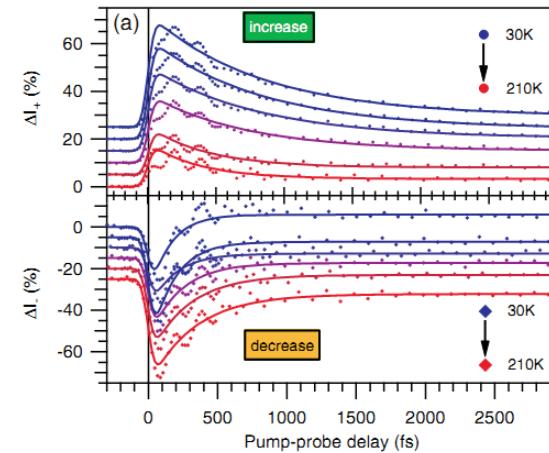
# TR-ARPES in the SDW state

$\text{EuFe}_2\text{As}_2$

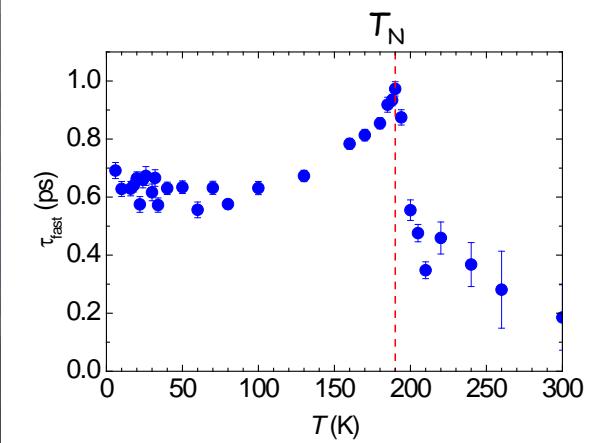
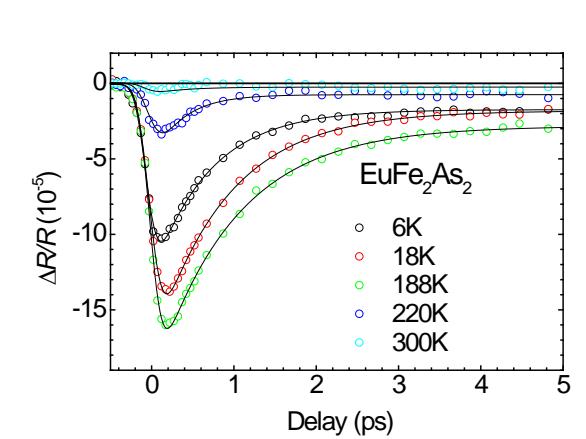


Rettig et. al., PRL 108, 097002 (2012)

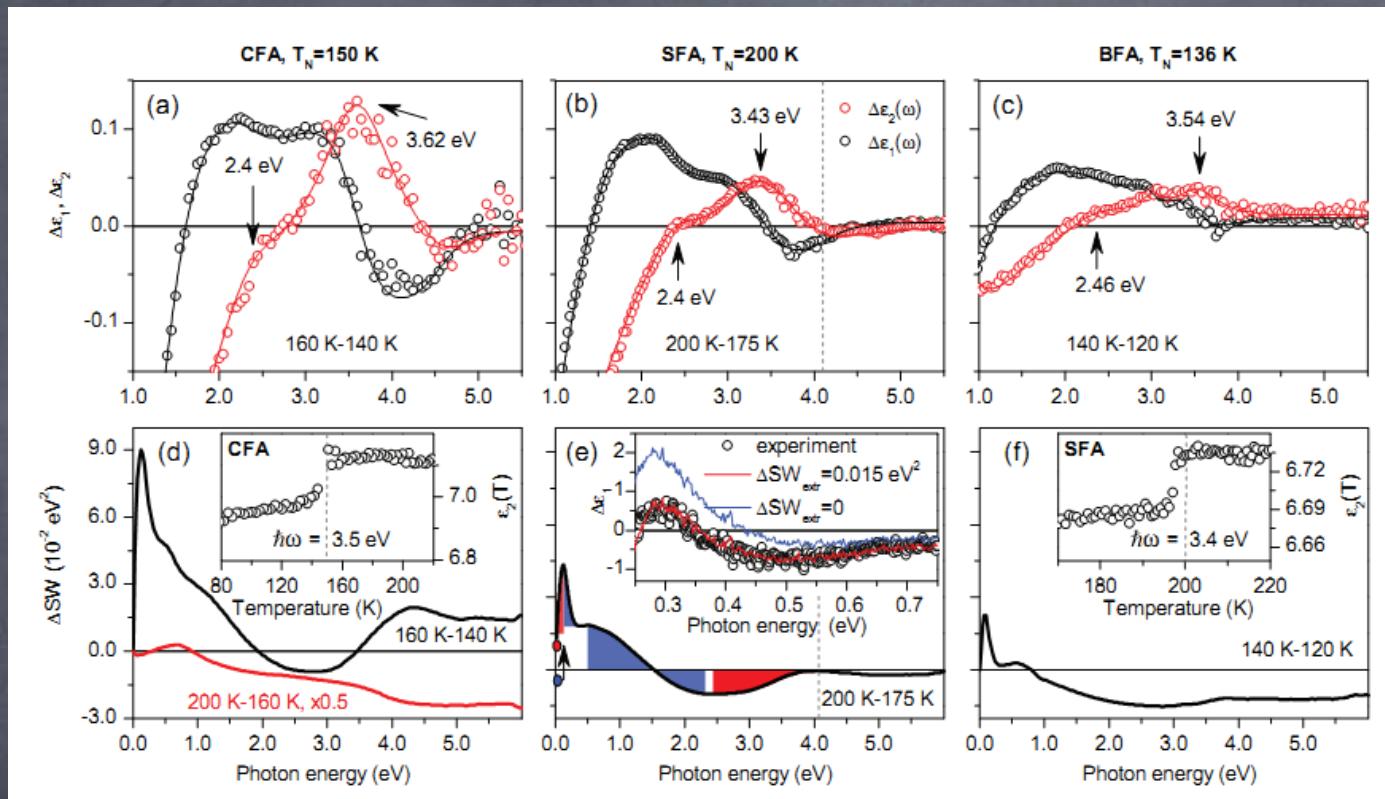
ARPES



ALL-optical



# Static optical response to the magneto-structural transition in AeFe<sub>2</sub>As<sub>2</sub> (Ae=Ba, Cs, Sr)

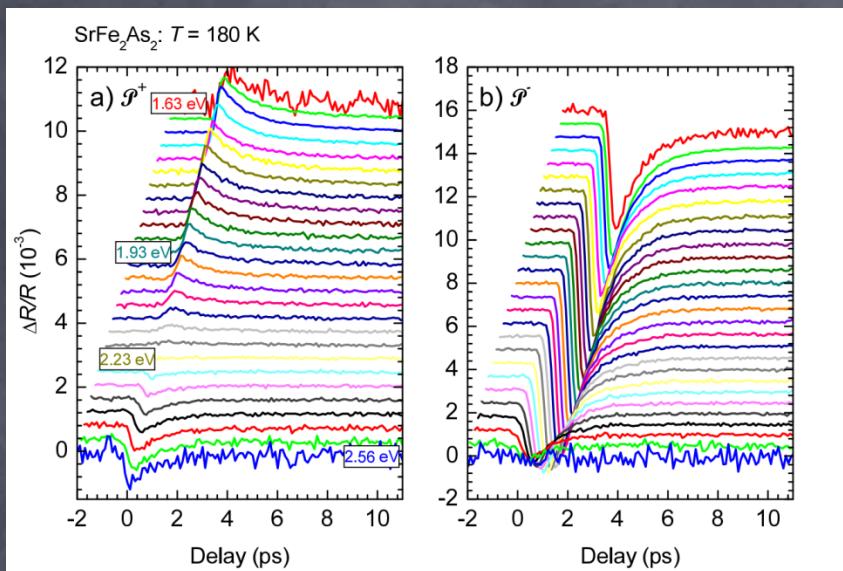


Charnukha et al., PRB **88**, 184511 (2013)

$$\varepsilon(\omega, T) = \varepsilon_0(\omega, T) + \frac{d\varepsilon(\omega)}{d\Delta} \Delta(T)$$

$$\delta R(\omega, t) \approx g_{QP}(\omega) n_{pe}(\omega, t) + \frac{dR(\omega)}{d\Delta} \delta\Delta(t) + \dots$$

# SDW state: spectral dependence

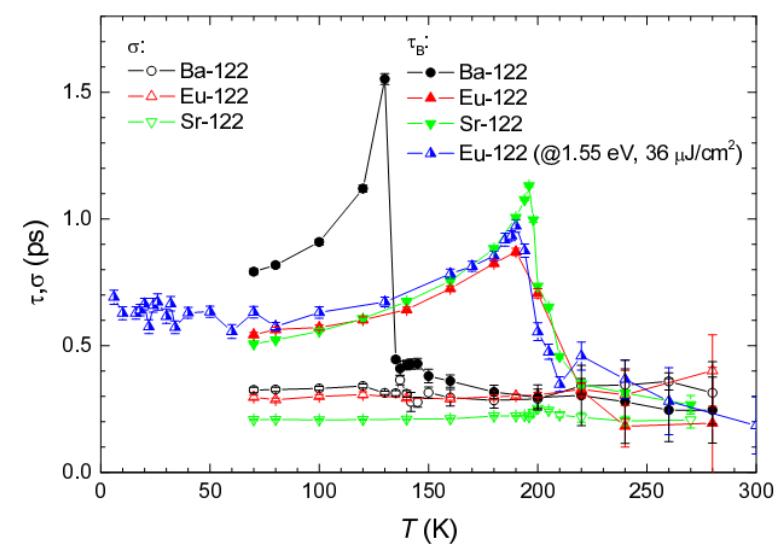
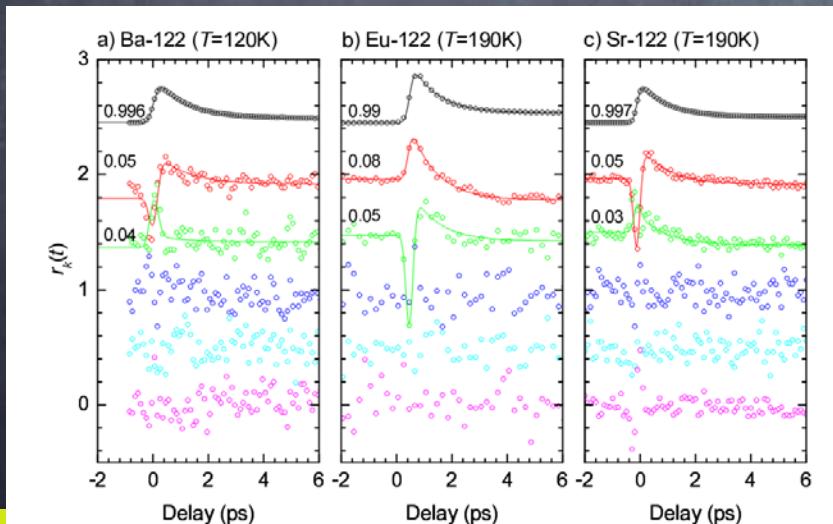


$$\Delta R_{\alpha,\beta} \propto \int d^3k [ |M_{\alpha,\beta}(\mathbf{k})|^2 \Delta f_{\alpha}(\mathbf{k}) \\ \times g(\epsilon_{\beta}(\mathbf{k}) - \epsilon_{\alpha}(\mathbf{k}) - \hbar\omega_{\text{probe}})].$$

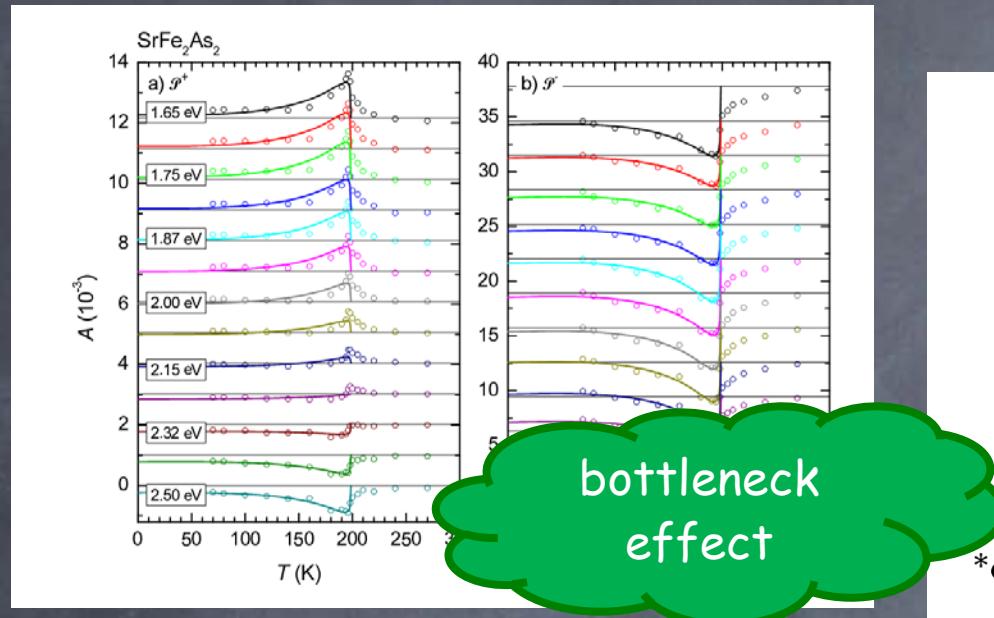
Singular value decomposition:

$$\left( \frac{\Delta R(t_j)}{R} \right) (\omega_{\text{pr},i}) = \sum w_k u_{ik} v_{jk} \\ = \sum A_k (\hbar\omega_{\text{pr},i}) r_k(t_j).$$

Pogrebna et al., PRB 89, 165131 (2014)



# Bottleneck model fits



sample	$2\Delta(0)/k_B T_{SDW}$	$g_{ph}$
Ba-122	$8 \pm 6$	$3.2 \pm 1.1$
Eu-122	$13 \pm 8$	$2.8 \pm 1.4$
Sr-122	$8 \pm 3$	$2.7 \pm 0.8$
$YBa_2Cu_3O_{7-\delta}^1$	10	$\sim 60$
*Ba-122	$7.7^{30}, \sim 9^{38}$	-
*Sr-122	$8.7^{30}, \sim 9^{38}$	-
*Eu-122	$5.6^{39}$	-

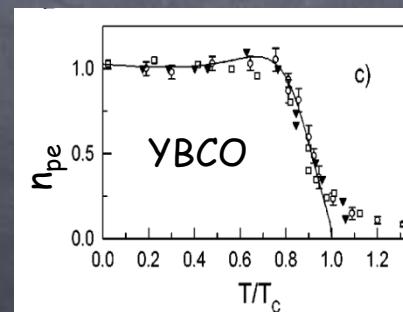
\*optical conductivity

\*Charnukha et al., PRB **88**, 184511 (2013); Hu et al., PRL **101**, 257005 (2008); Wu et al., PRB **79**, 155103 (2009)

$$\Delta R = \left[ \gamma_0 + \eta \frac{\Delta^2(T)}{\Delta^2(0)} \right] n_{pe}$$

$$n_{pe} \propto 1 / \left[ \left( \frac{2\Delta(T)}{k_B T_{SDW}} + \frac{T}{T_{SDW}} \right) \left( 1 + g_{ph} \sqrt{\frac{k_B T}{\Delta(T)}} \exp \left( -\frac{\Delta(T)}{k_B T} \right) \right) \right]$$

$\Delta$   
 $g_{ph} - \epsilon_F$   
matrix element  
effect  
involved bosons



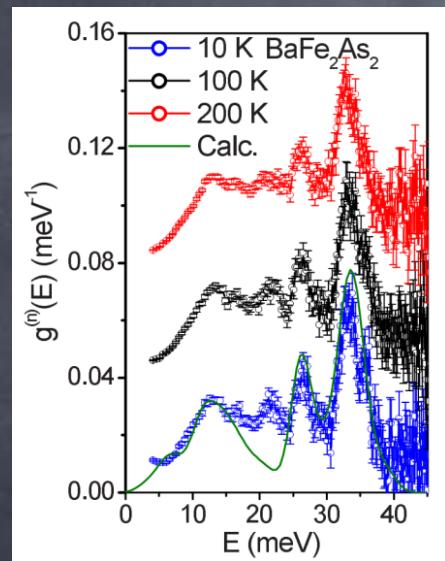
V.V. Kabanov et al., PRB **59**, 1497 (1999)

Pogrebna et al., PRB **B 89**, 165131 (2014)

# Bottleneck boson?

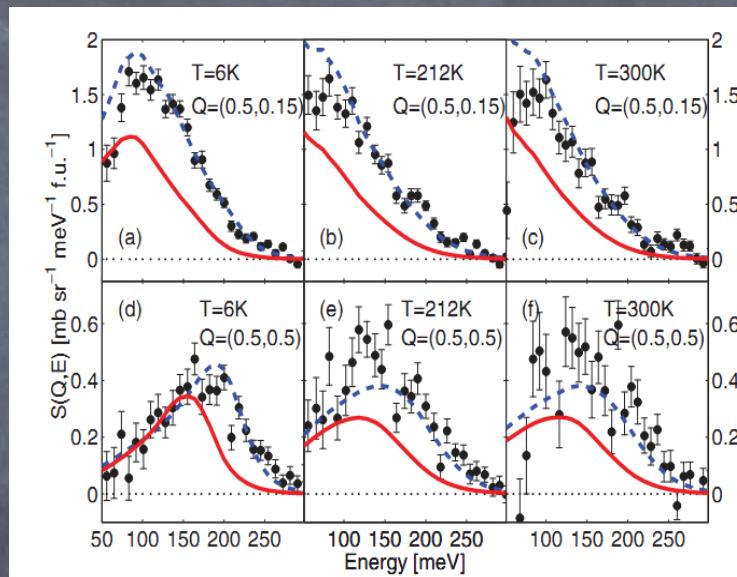
$$2\Delta_{SDW}(0) = \sim 100-200 \text{ meV}$$

phonons:



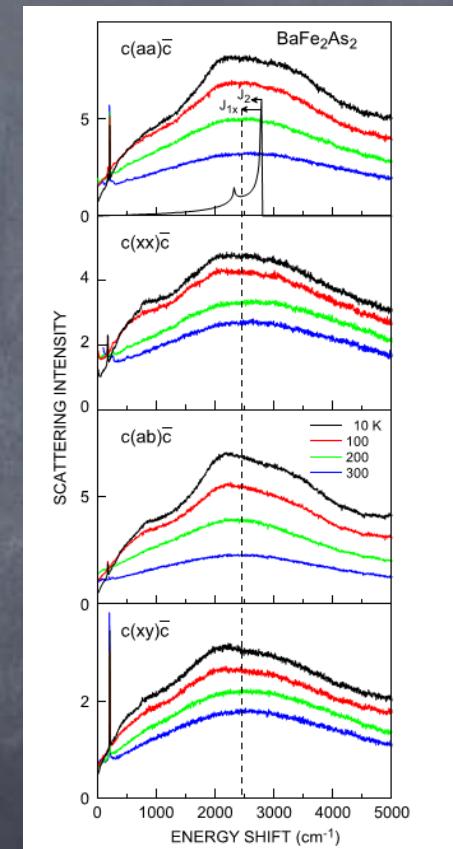
Mittal et al.,  
PRB 78, 104514 (2008)

magnons:



Ewings et al., PRB 83, 214519 (2011)

Magnons have a proper energy scale to play  
the role of the bottleneck boson.

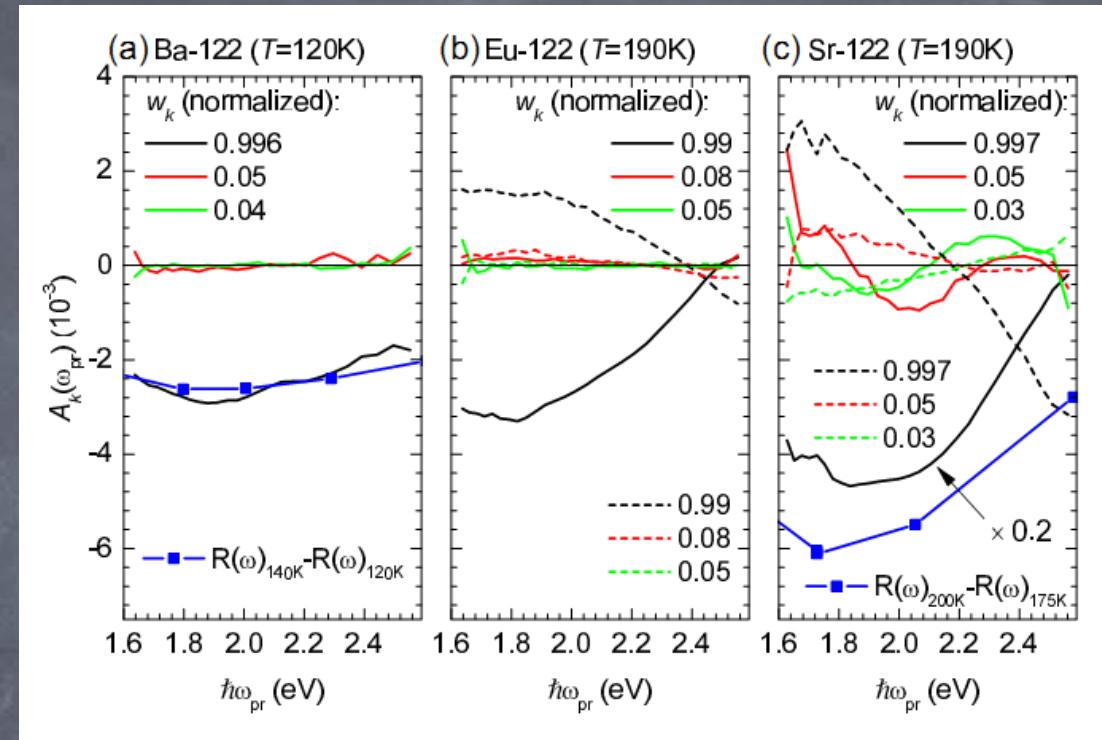


S. Sugai et al.,  
J. Phys. Soc. Jpn. 81,  
024718 (2012)

# Spectral dependence, comparison to equilibrium response

Equilibrium:

$$R|_{T>T_N} - R|_{T<T_N} \sim \frac{dR}{d\Delta} \Delta(T)$$



Transient:

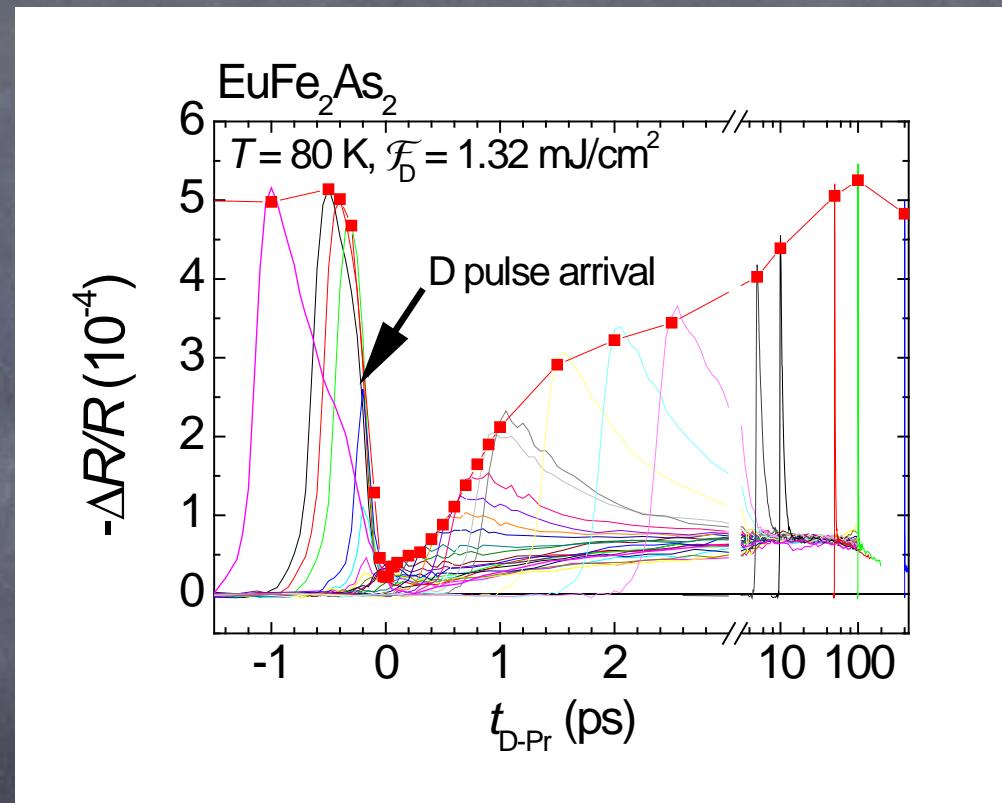
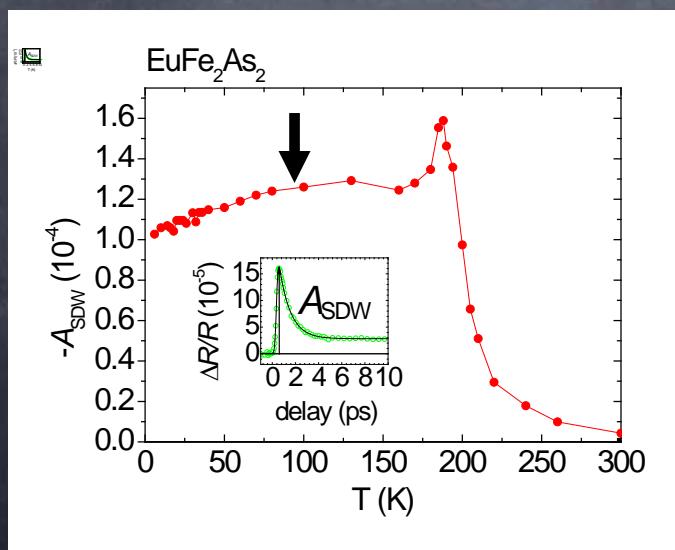
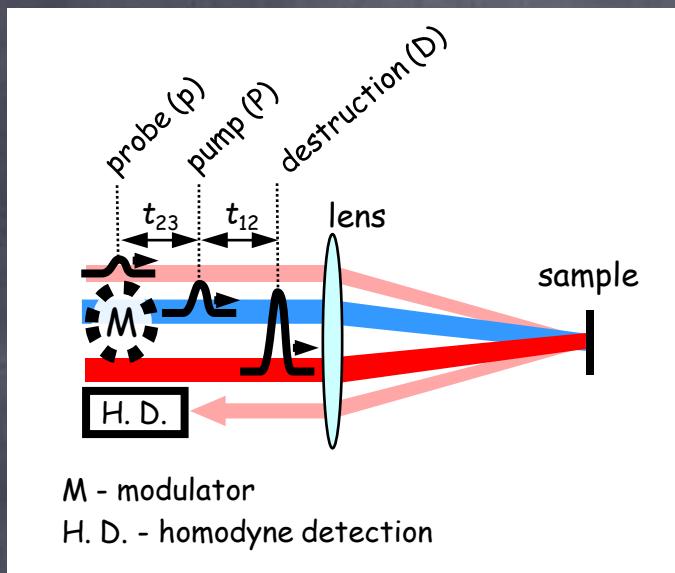
quasiparticle:  $\delta R(t) \propto \int d^3k |M(\mathbf{k})|^2 g(\varepsilon_\beta - \varepsilon_\alpha - \hbar\omega) \delta f(\mathbf{k}, t) \sim c_0(T, \Delta) n_{pe}(t)$

collective:  $\delta R(t) \propto \frac{dR}{d\Delta} \delta\Delta(t) \sim c_1(T, \Delta) n_{pe}(t)$

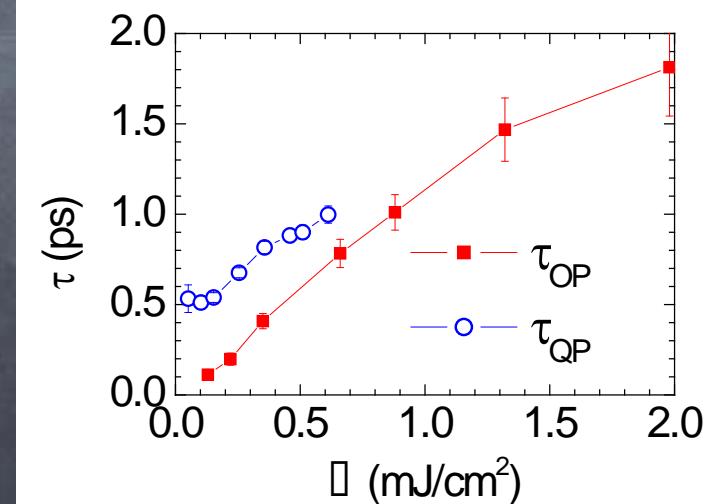
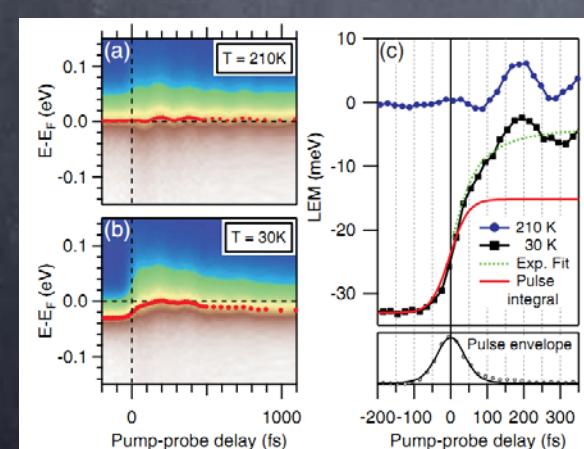
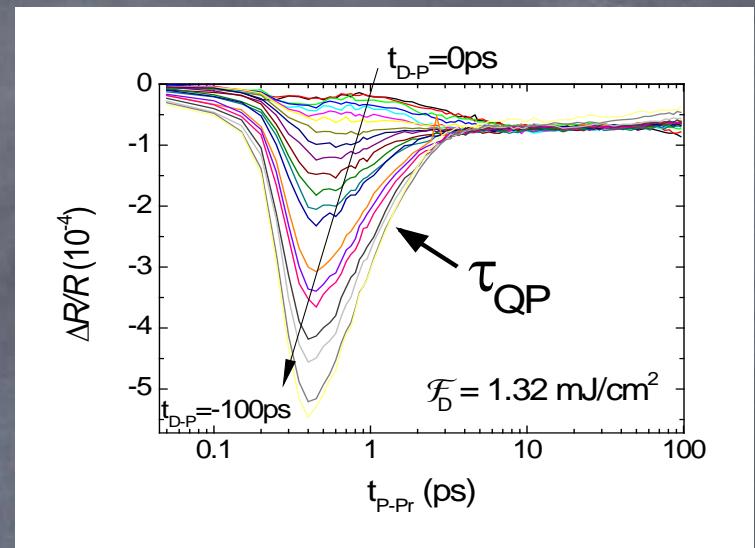
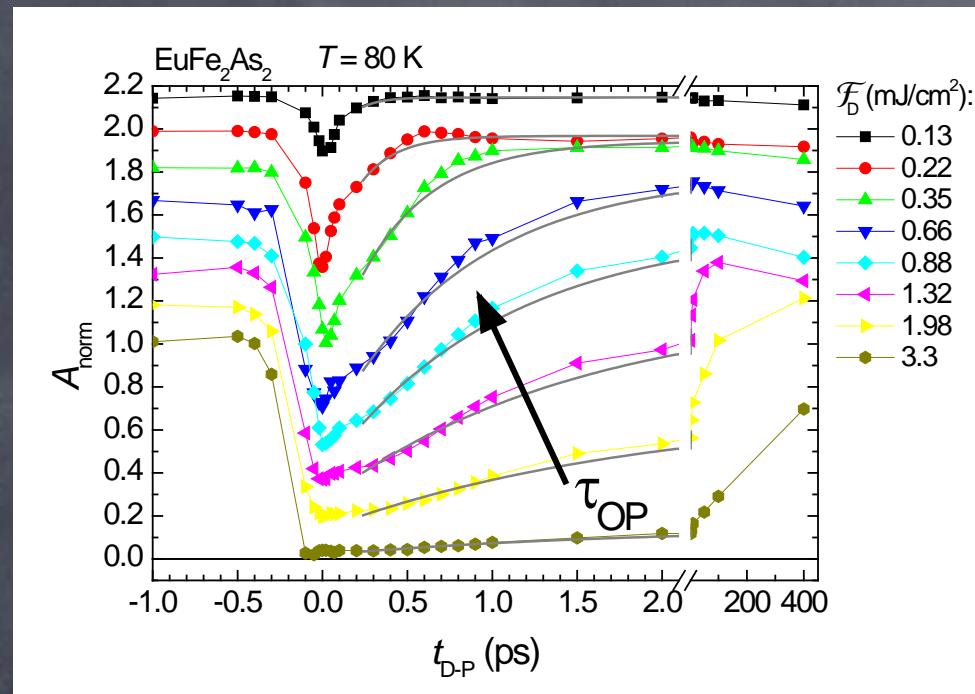
gap eq.:  $\Delta = \sum V \frac{\Delta}{\sqrt{\Delta^2 + \varepsilon_{\mathbf{k}}^2}} [1 - 2 f(\mathbf{k}')] \Rightarrow$  linear response:  $\delta\Delta(t) \propto n_{pe}(t)$

amplitude mode:  $\hbar\omega_A = \sqrt{(2\Delta)^2 + (v_F q)^2} \cdot \frac{1}{\omega_A} = \frac{\hbar}{2\Delta} \sim 3 \text{ fs}$

# Multiple pump pulse technique



# Order parameter dynamics upon quench



# Conclusions

- CDWs: Rothwarf-Taylor bottleneck is possible just near the transition when the gap is still small. The all-optical transient response is not yet fully understood. The role of an overdamped collective electronic mode?
- SDW state of iron based pnictides: bottleneck governed relaxation dynamics at weak excitation.
- High energy magnons as the bottleneck bosons.
- Faster order parameter dynamics observed at higher excitation possibly driven by coherent SRS excitation of the order parameter.