Bypassing the Structural Bottleneck in the Ultrafast Melting of Electronic Order

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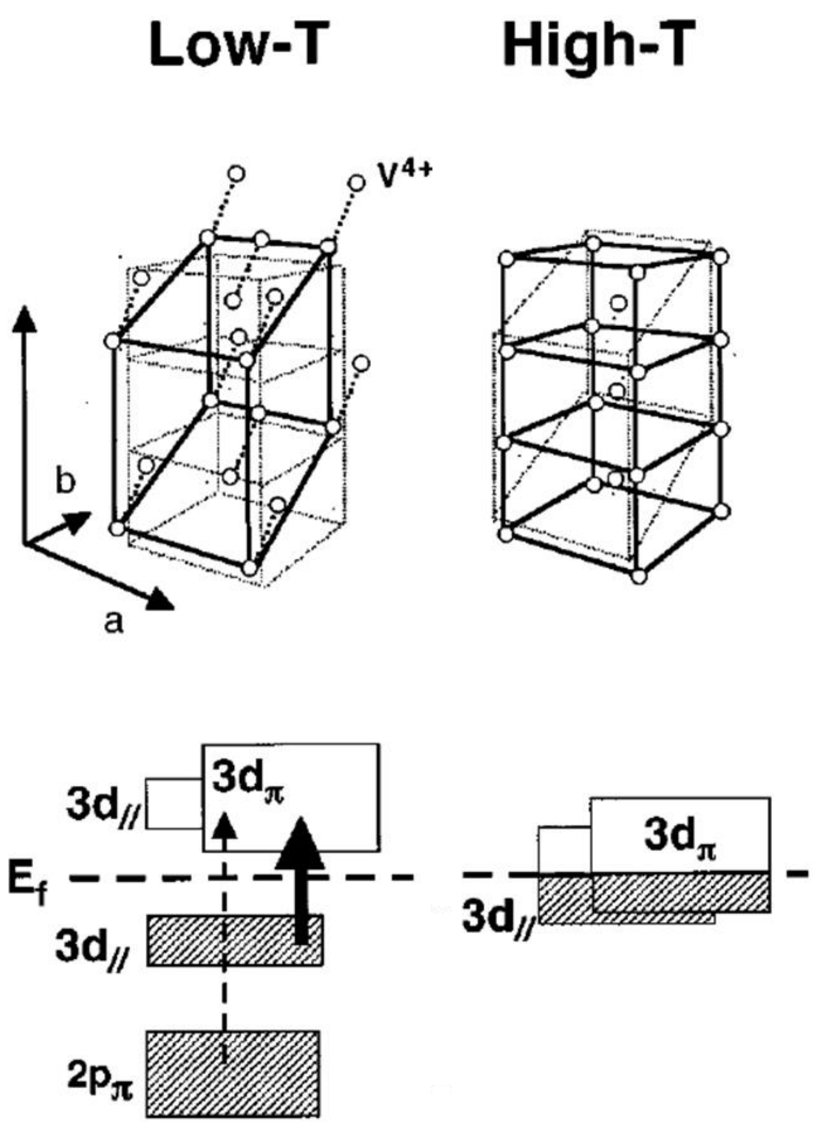
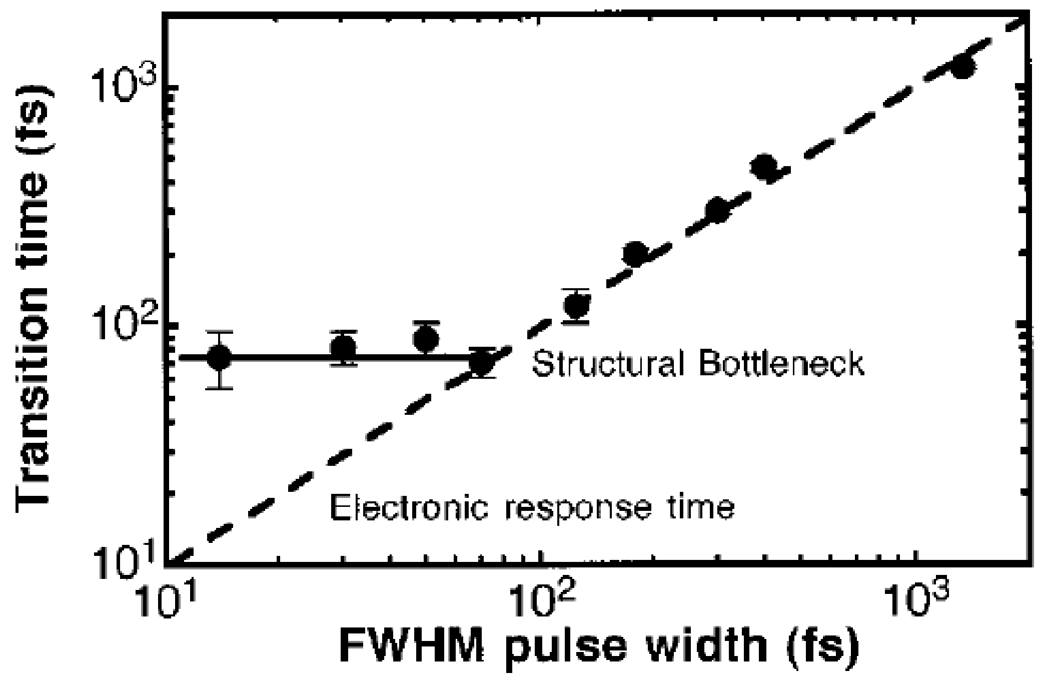
http://www.ieap.uni-kiel.de/solid/ag-bauer/

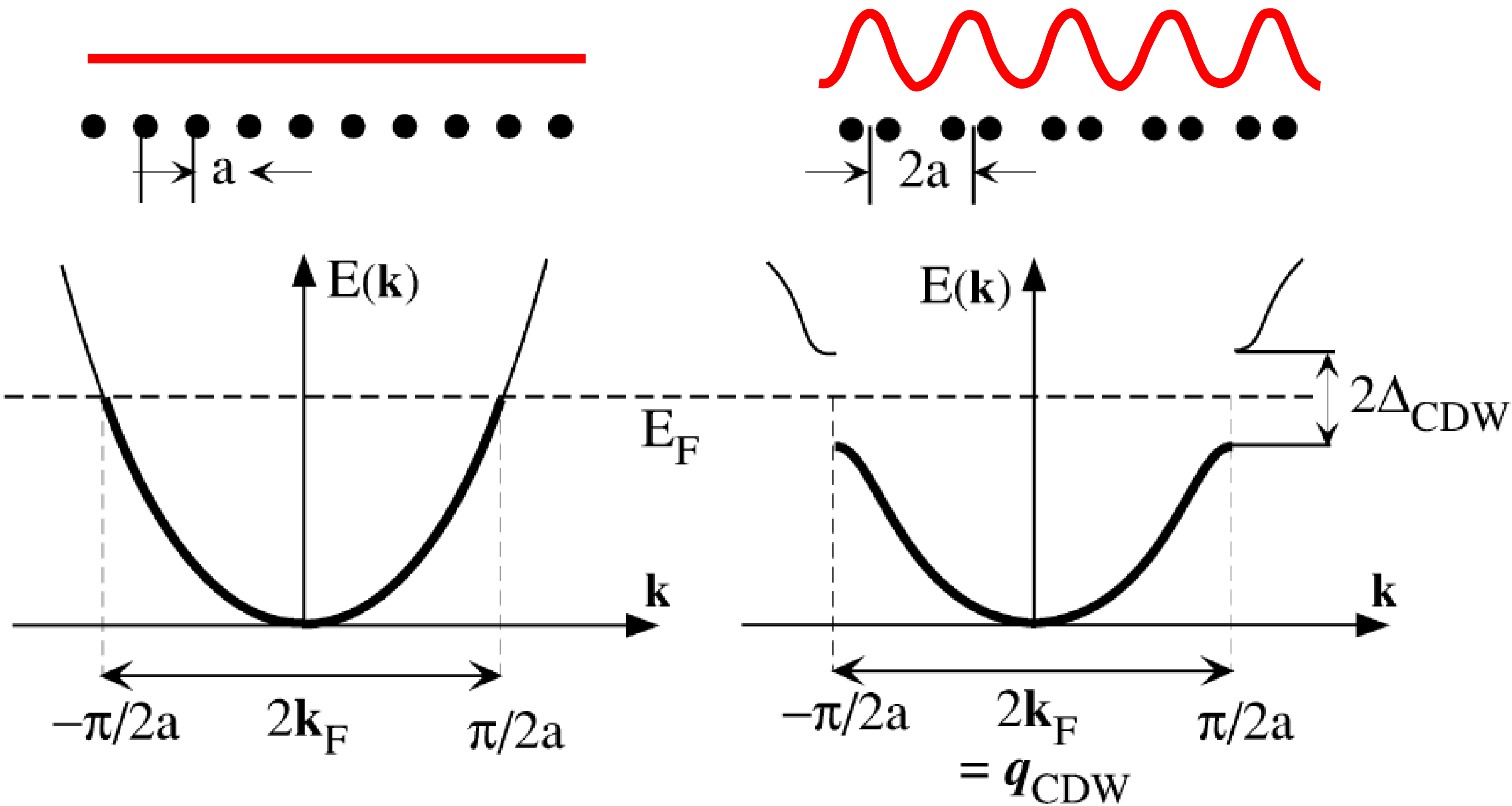
Insulator-to-metal transition in VO_2





A. Cavalleri, et al.,
Phys. Rev. B 70, 161102(R) (2004)

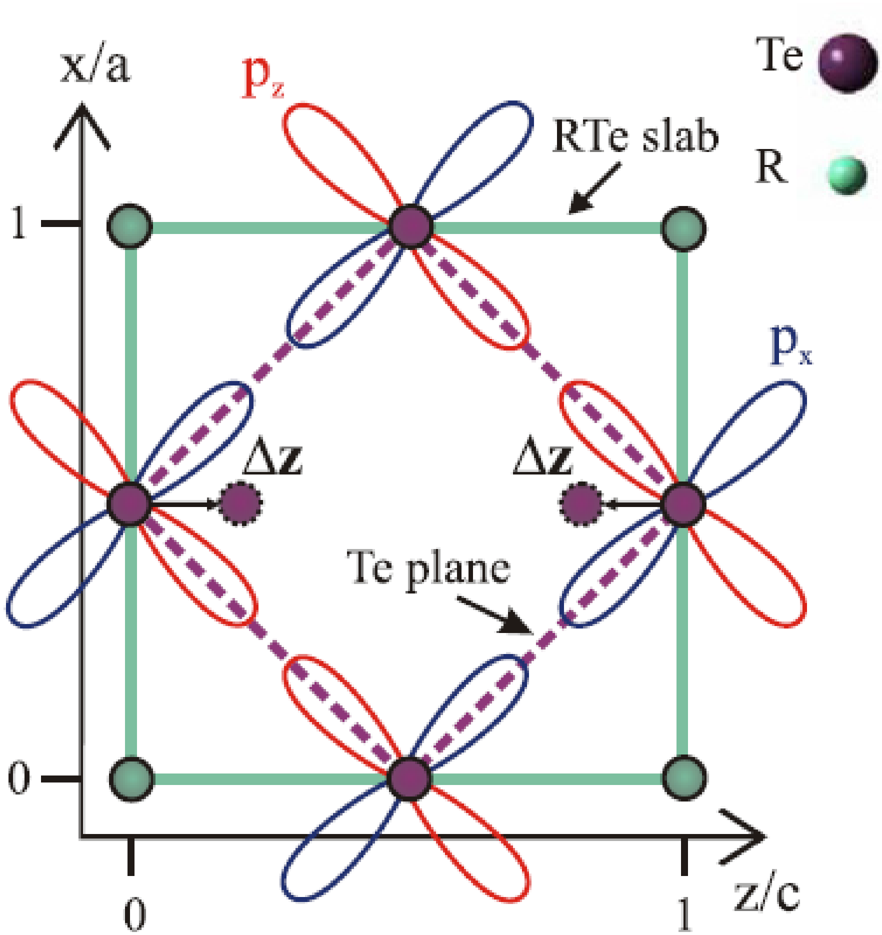
Peierls instability



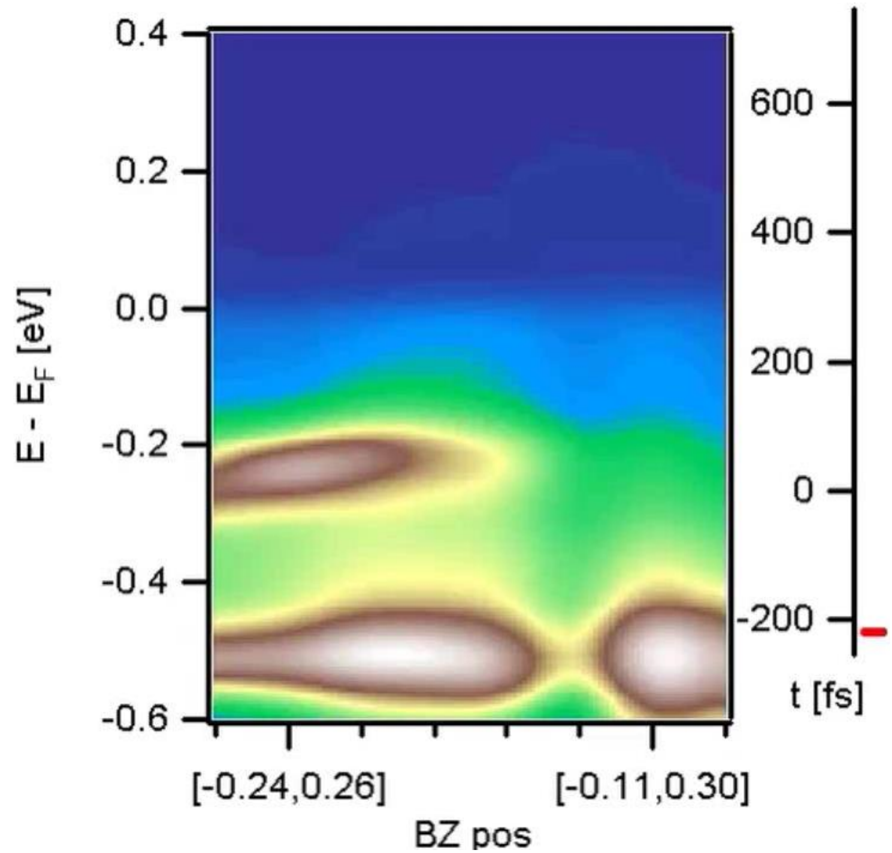
R. Peierls, Quantum Theory of Solids, Clarendon, Oxford (1955)

CDW Melting in $TbTe_3$

lattice



(transient) electronic structure



F. Schmitt et al., Science 321, 1649 (2008)
P. Kirchmann et al., NJP 13, 063022 (2011)

outline

structural bottleneck ⇨ *coherent* lattice excitation

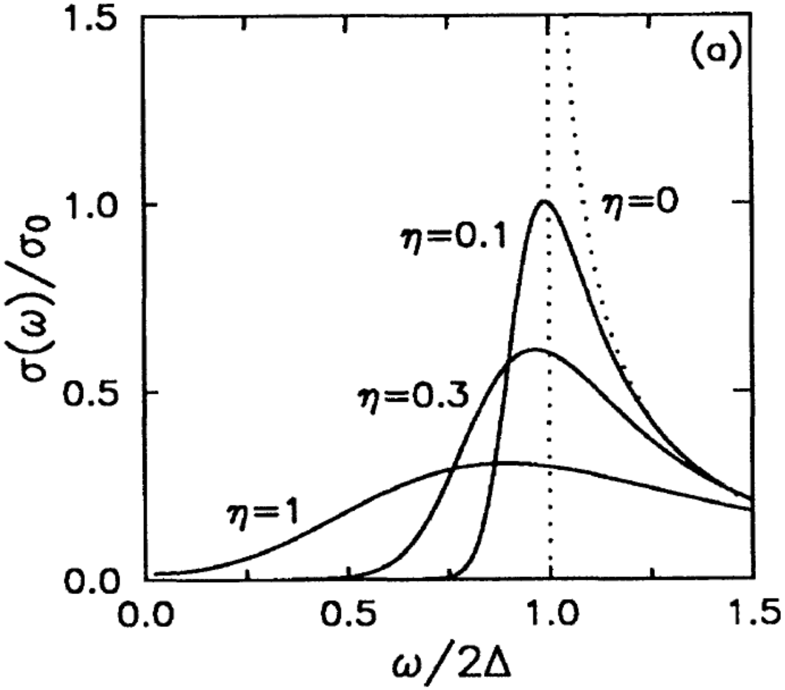
what‘s about *incoherent* lattice excitations?

- can they affect a symmetry-broken phase?
- what are the characteristic time-scales?

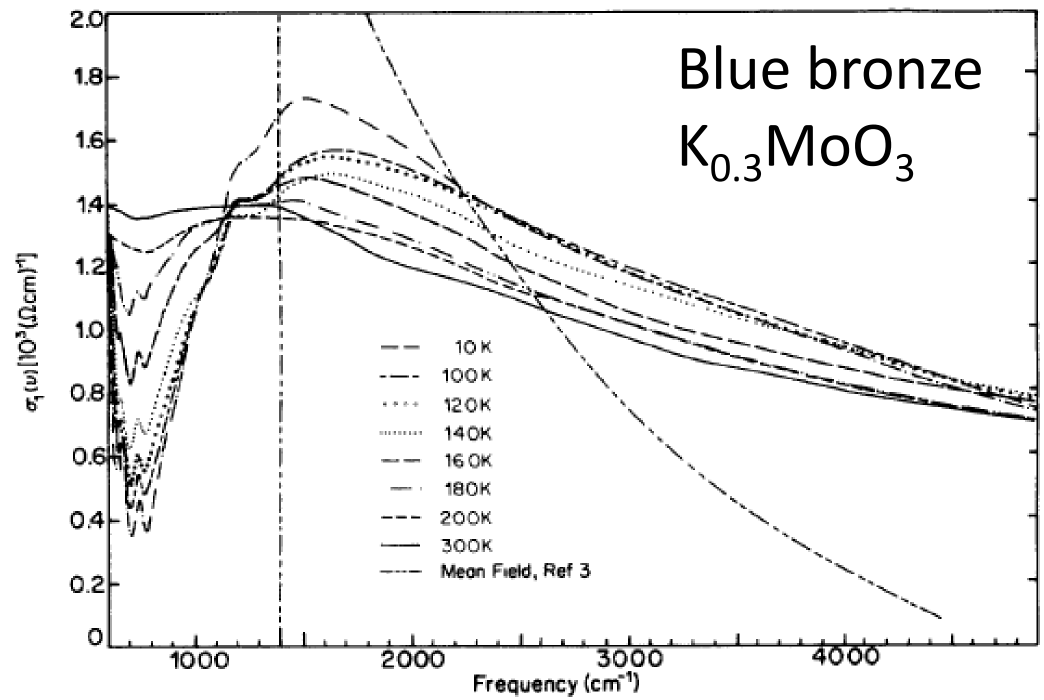
CDW Suppression by Lattice Fluctuations

1d Peierls System: effect of vibrational disorder

Theory



Experiment



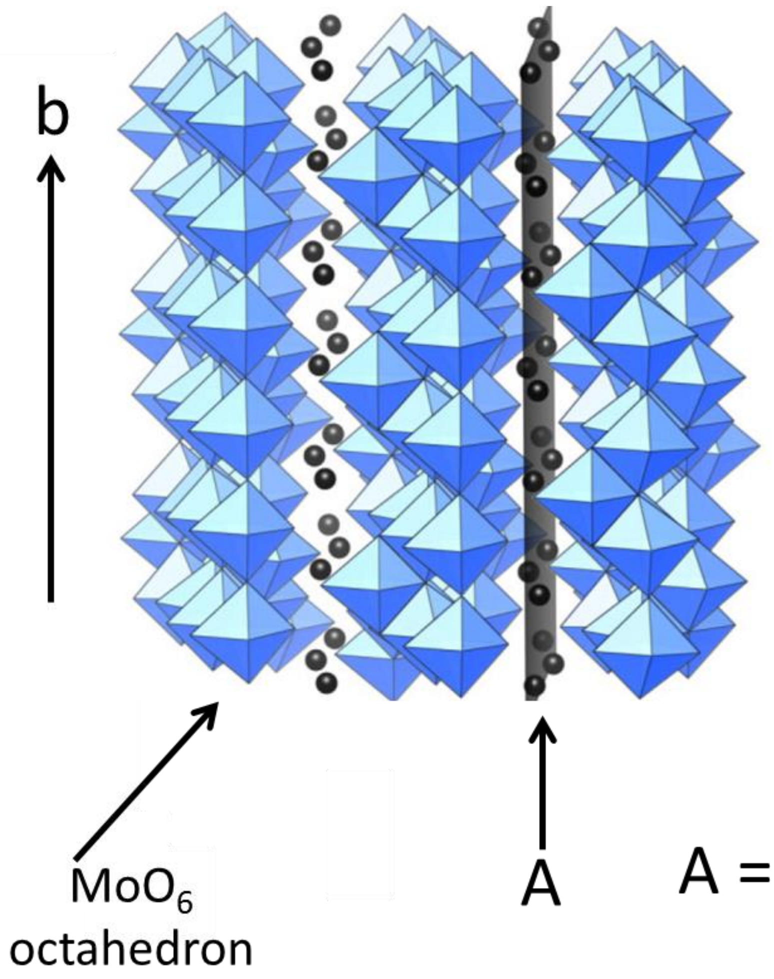
relevance of phase phonon modes
fluctuations: 3.7 – 99 meV

K. Kim, et al.,
Phys. Rev. Lett. 71, 4015 (1993)

L. Degiorgi, et al.,
Phys. Rev. B RC 49, 14754 (1994)

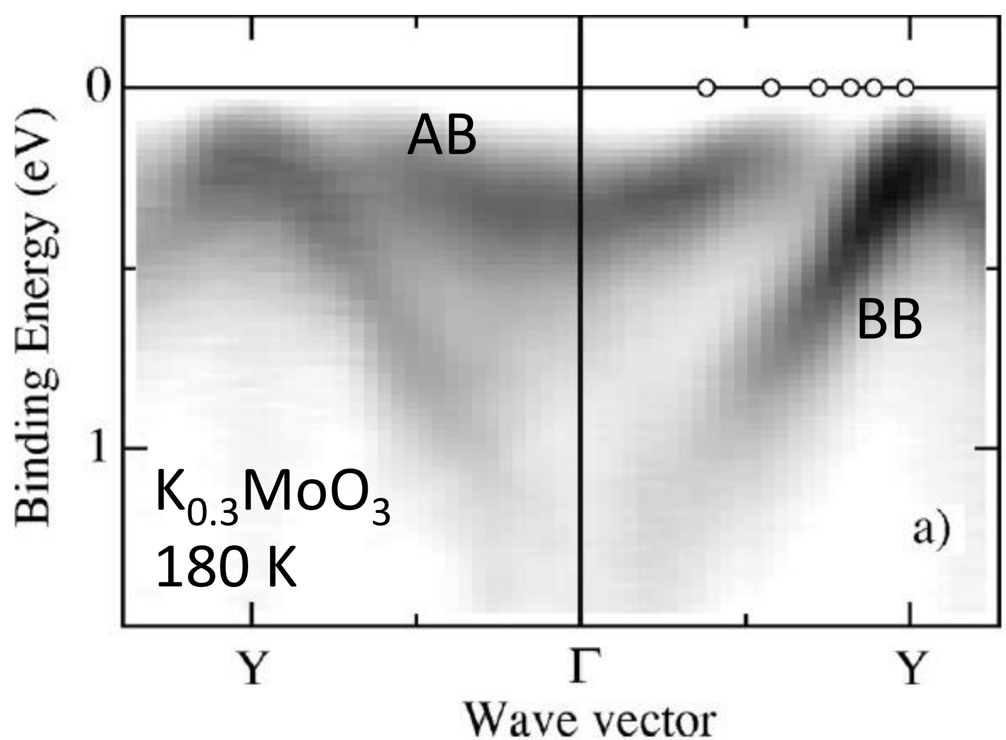
Prototypical Peierls insulator – A_xMoO_3

crystalline structure



A = K, Rb

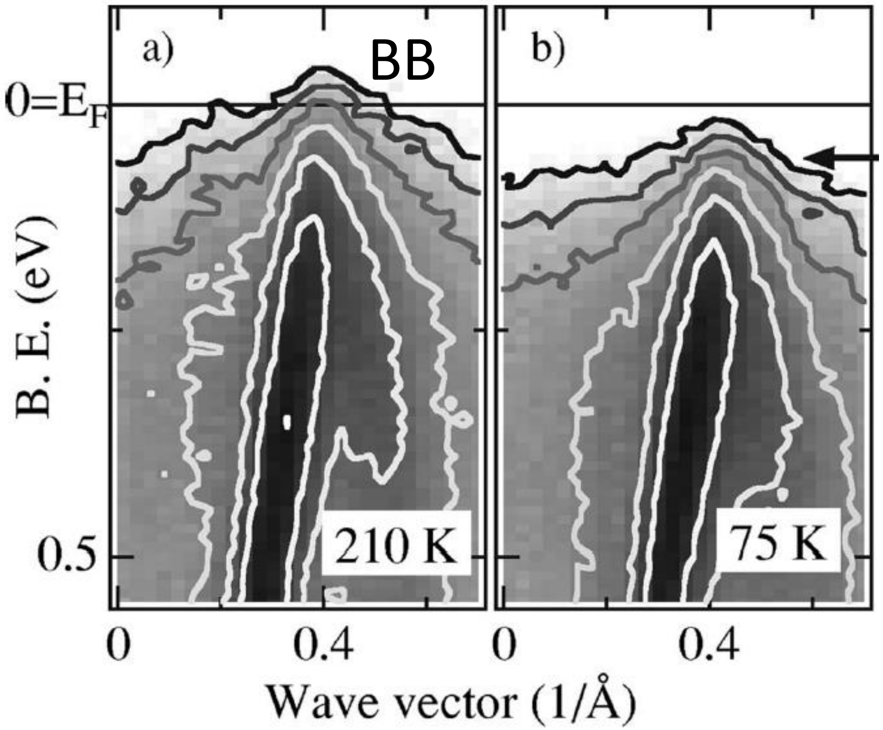
electronic structure
ARPES along b

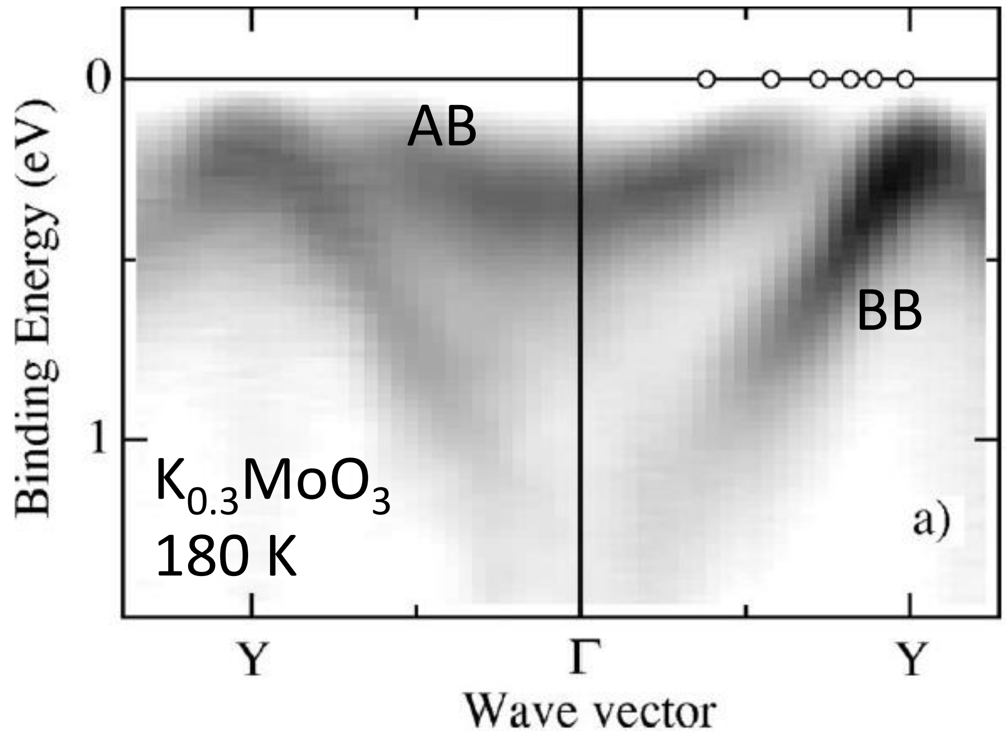


Perfetti *et al.*, PRB **66**, 075107 (2002)

Prototypical Peierls insulator – A_xMoO_3

electronic structure
ARPES along b

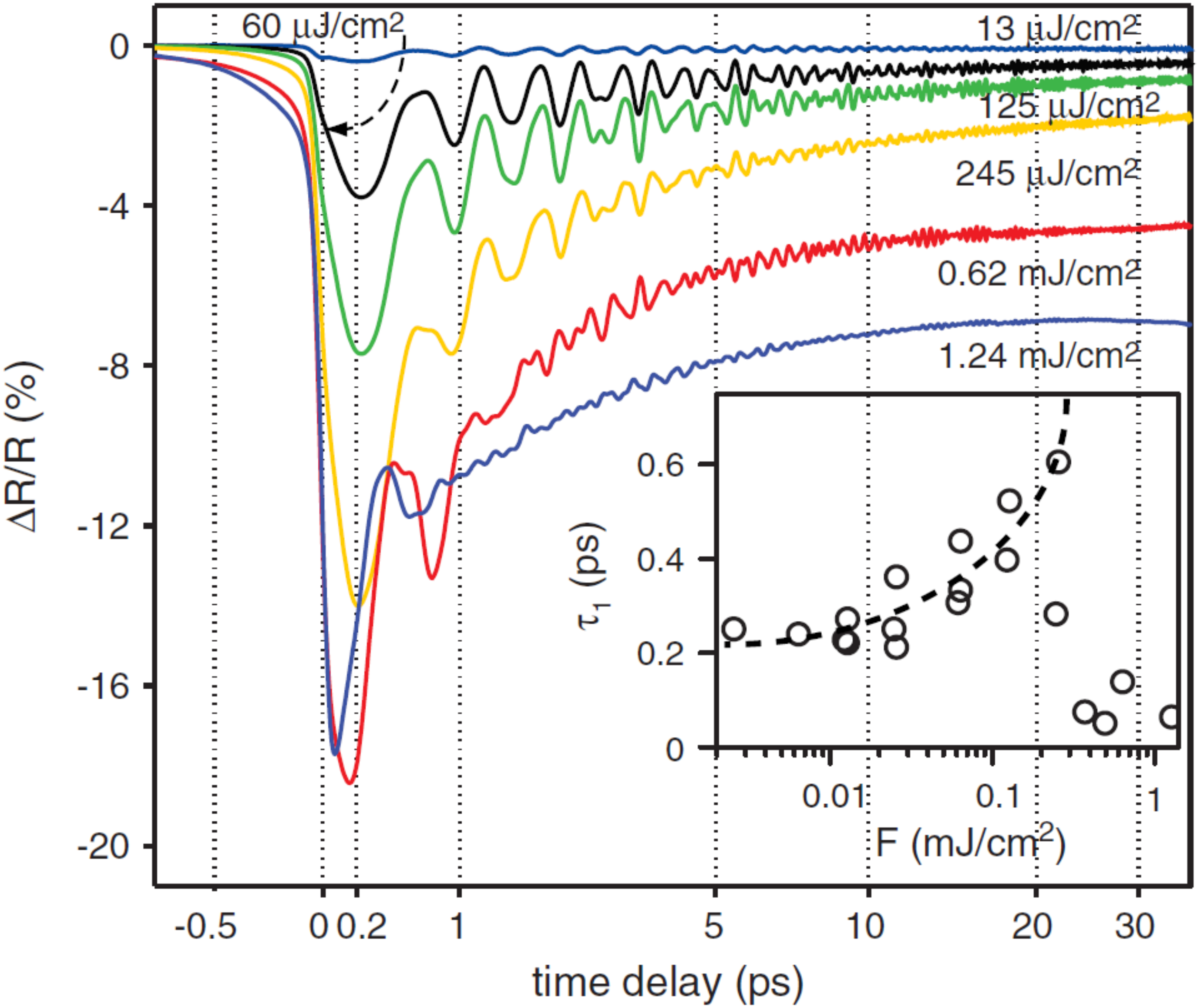




Perfetti *et al*., PRB **66**, 075107 (2002)

Prototypical Peierls insulator – A_xMoO_3

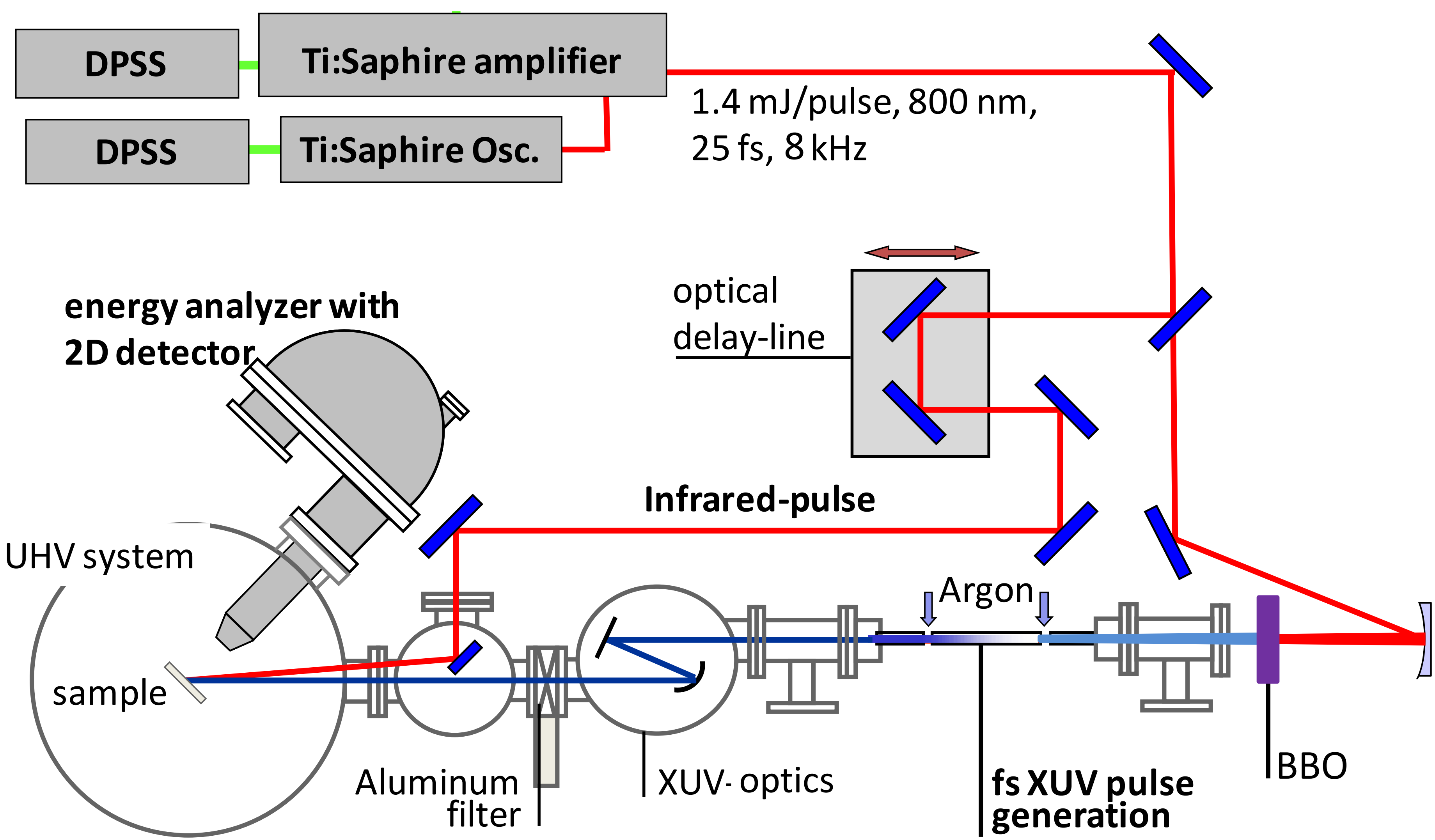
$K_{0.3}MoO_3$



(strongest) AM mode $T_{AM} \approx 600$ fs

Tomeljak *et al.*, PRL **102**, 066404 (2009)

HHG-based time-resolved ARPES

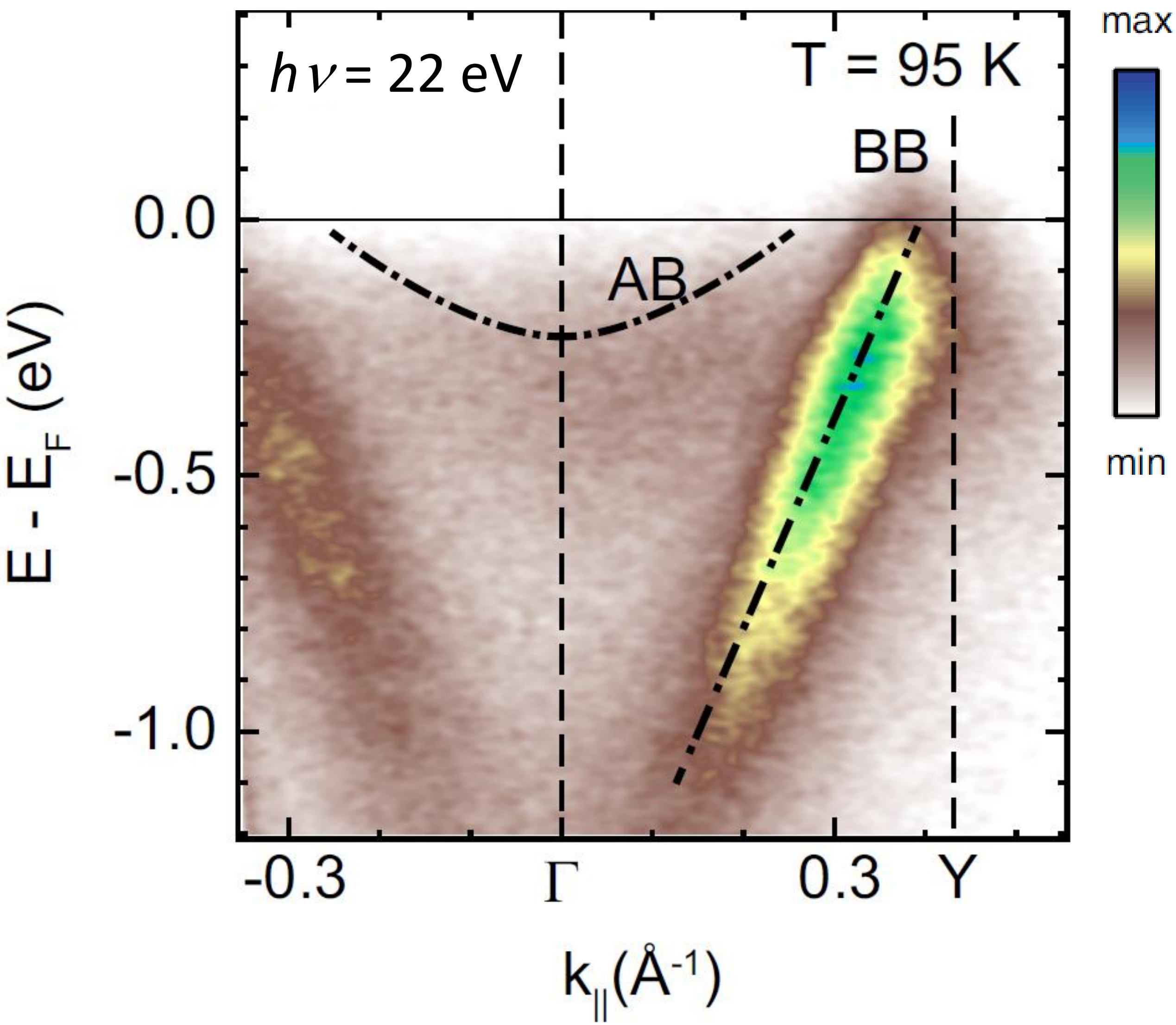


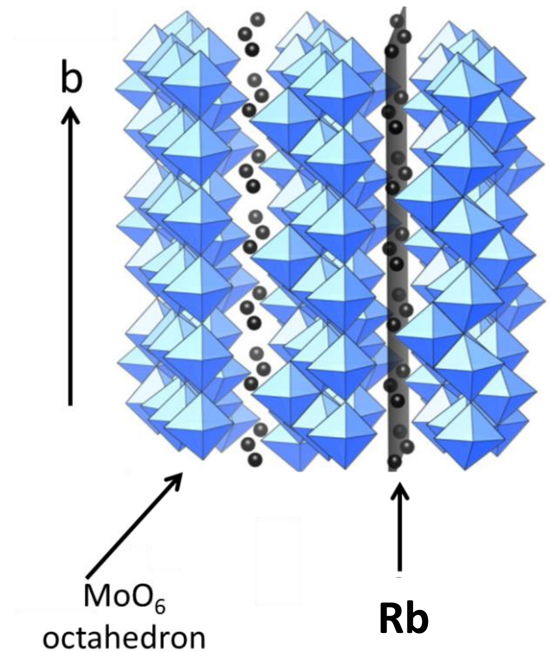
S. Mathias et al.; Rev. Sci. Instr. **78**, 083105 (2007)

S. Eich, et al.; J. Electr. Spect. Rel. Phen. (2014)

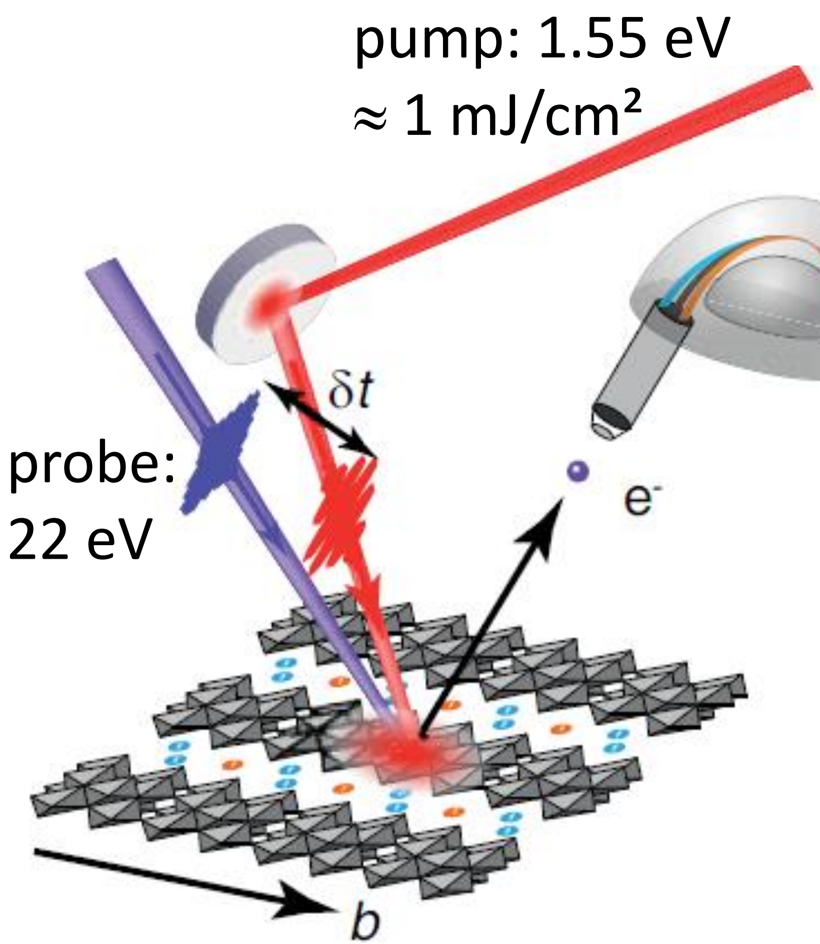
$Rb_{0.3}MoO_3$ – HHG-ARPES

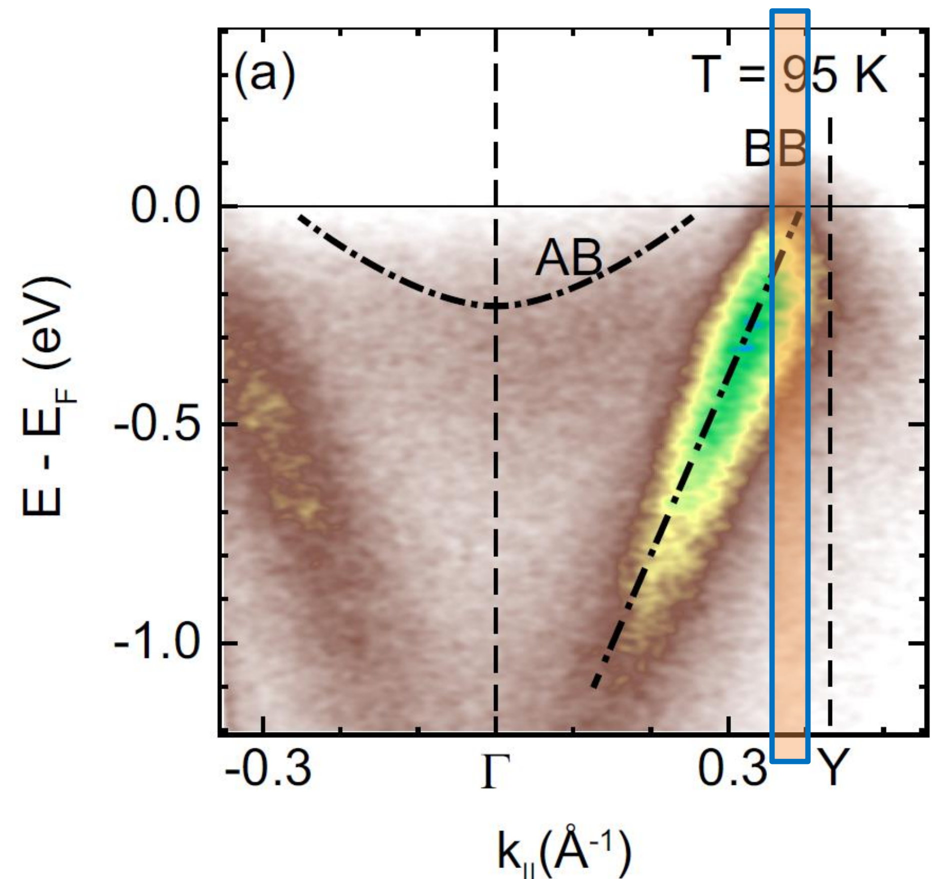
electronic structure along b





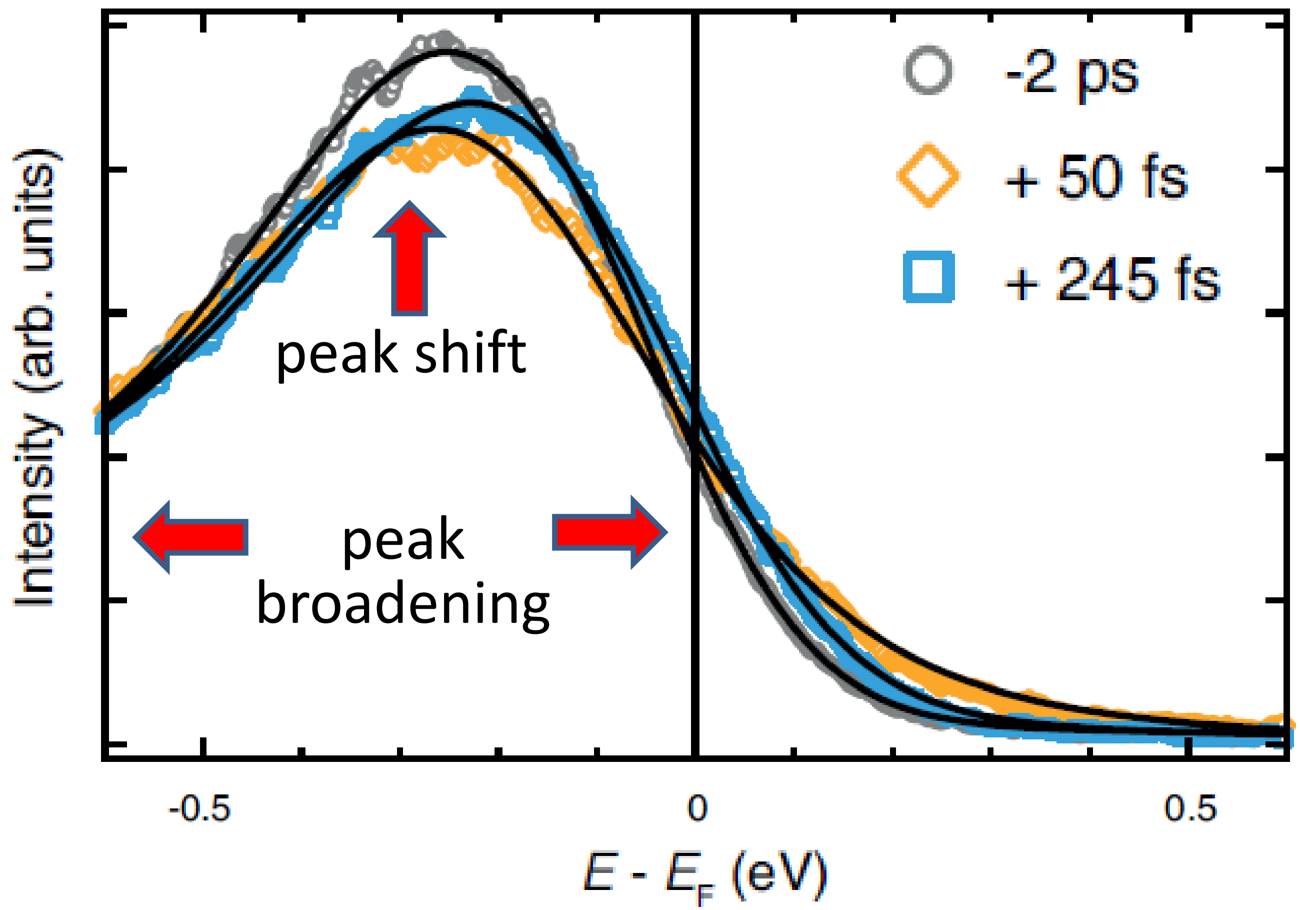
$Rb_{0.3}MoO_3$ – time-resolved ARPES





$Rb_{0.3}MoO_3$ – data analysis

EDC at k_F
95 K (CDW)
0.72 mJ/cm², 800 nm



Fit: Lorentzian ⊗ Fermi-Dirac ⊗ exp. Resolution

⇨ peak energy

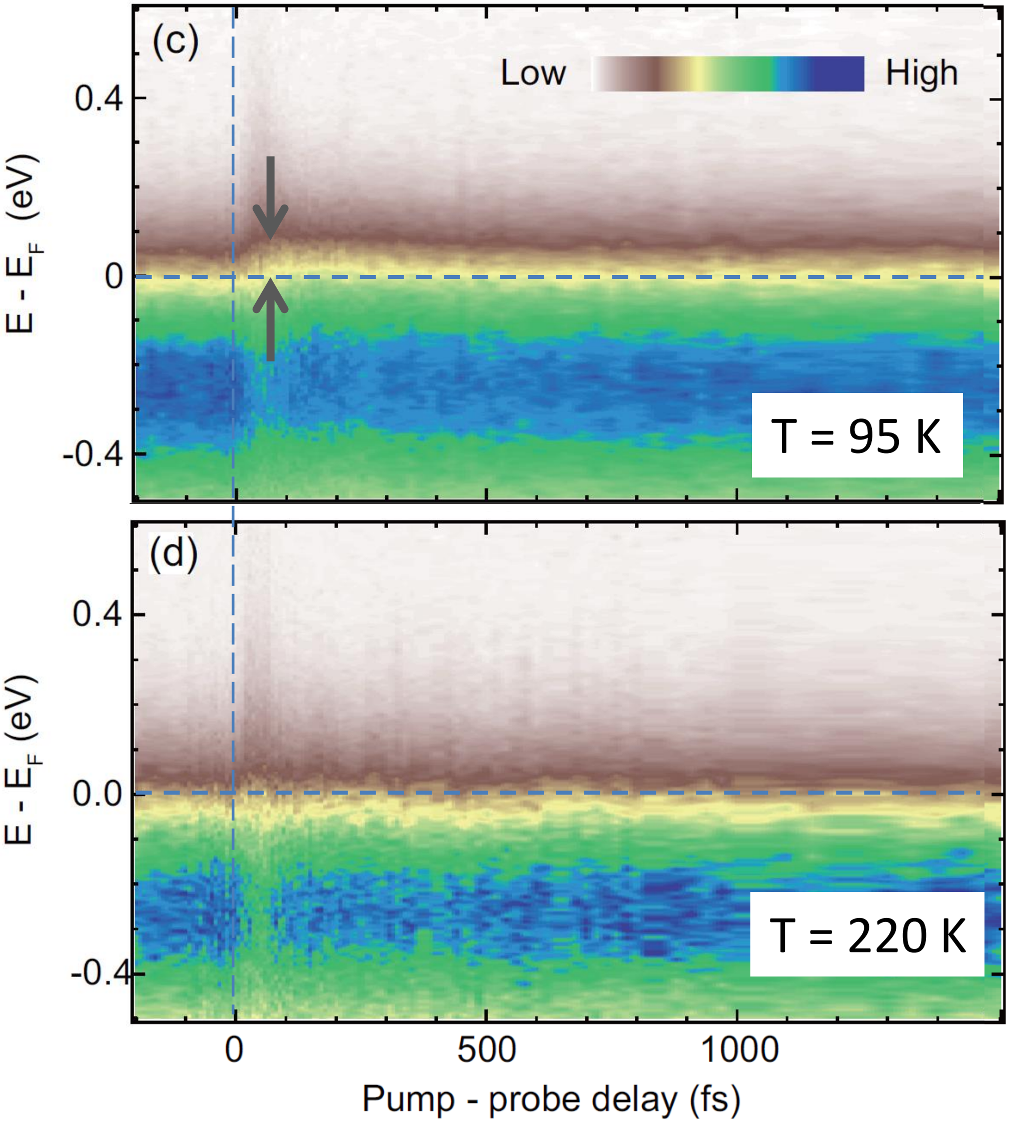
⇨ peak width

⇨ electron temperature

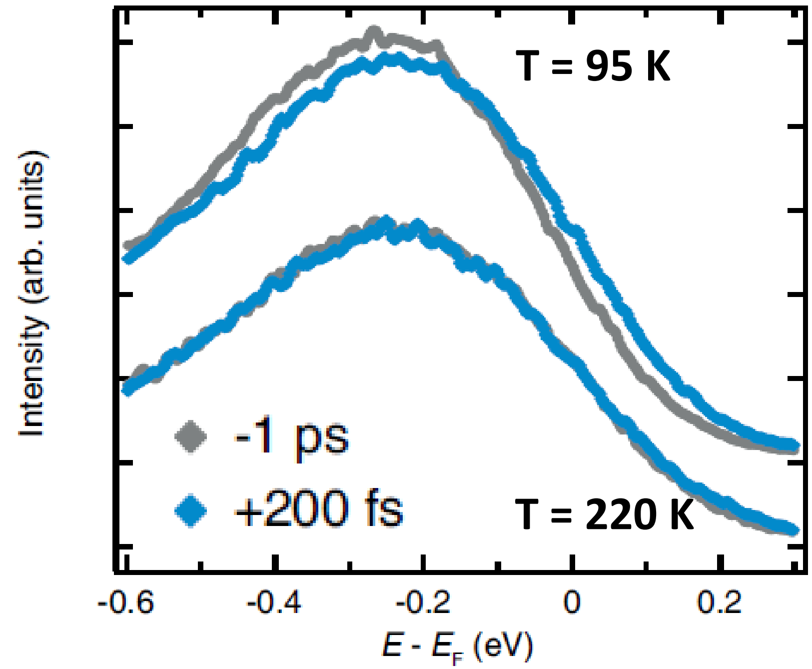
$Rb_{0.3}MoO_3$ – time-resolved ARPES

Is this gap closing?

0.72 mJ/cm²

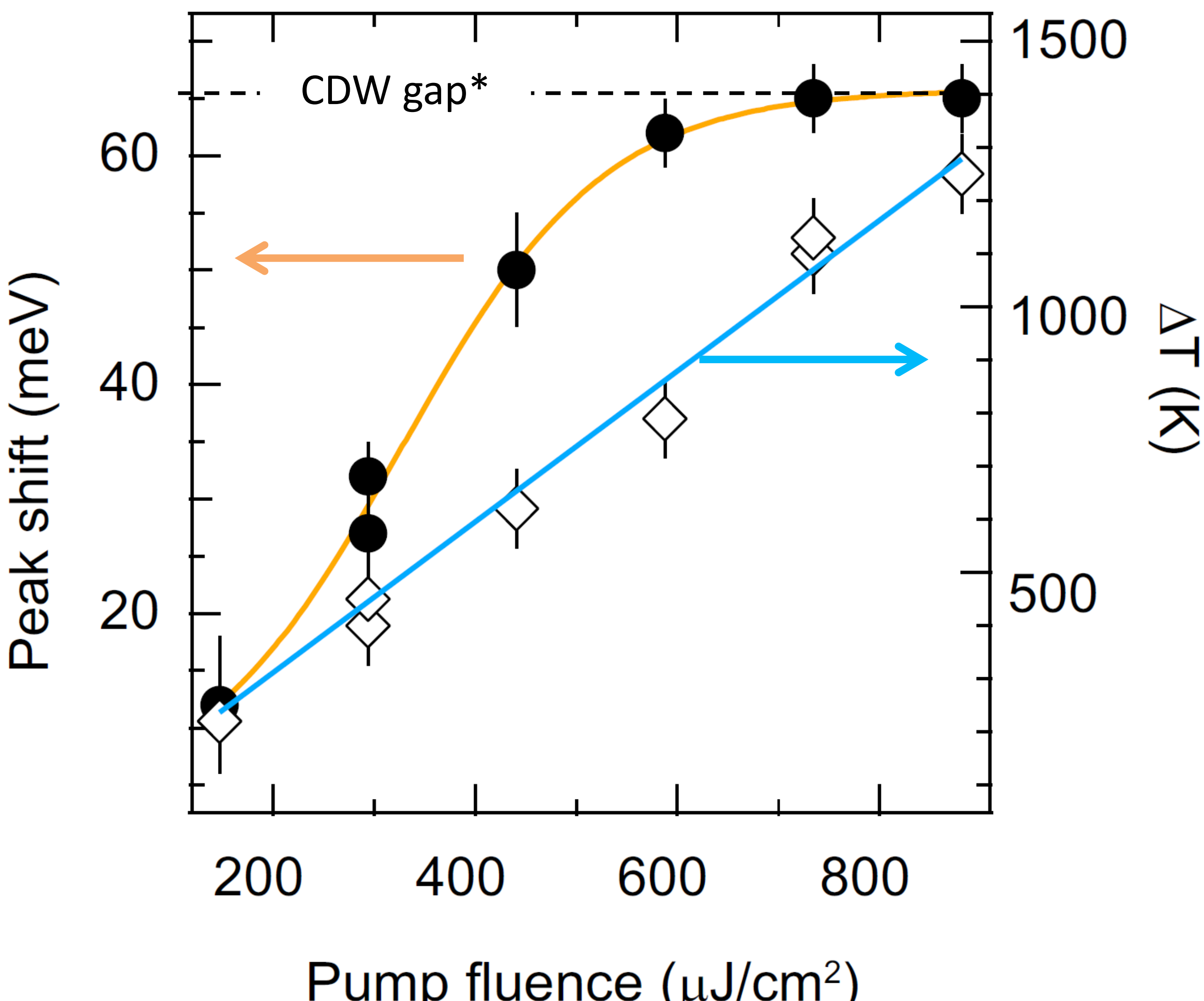


EDC at 0.35 ΓΥ



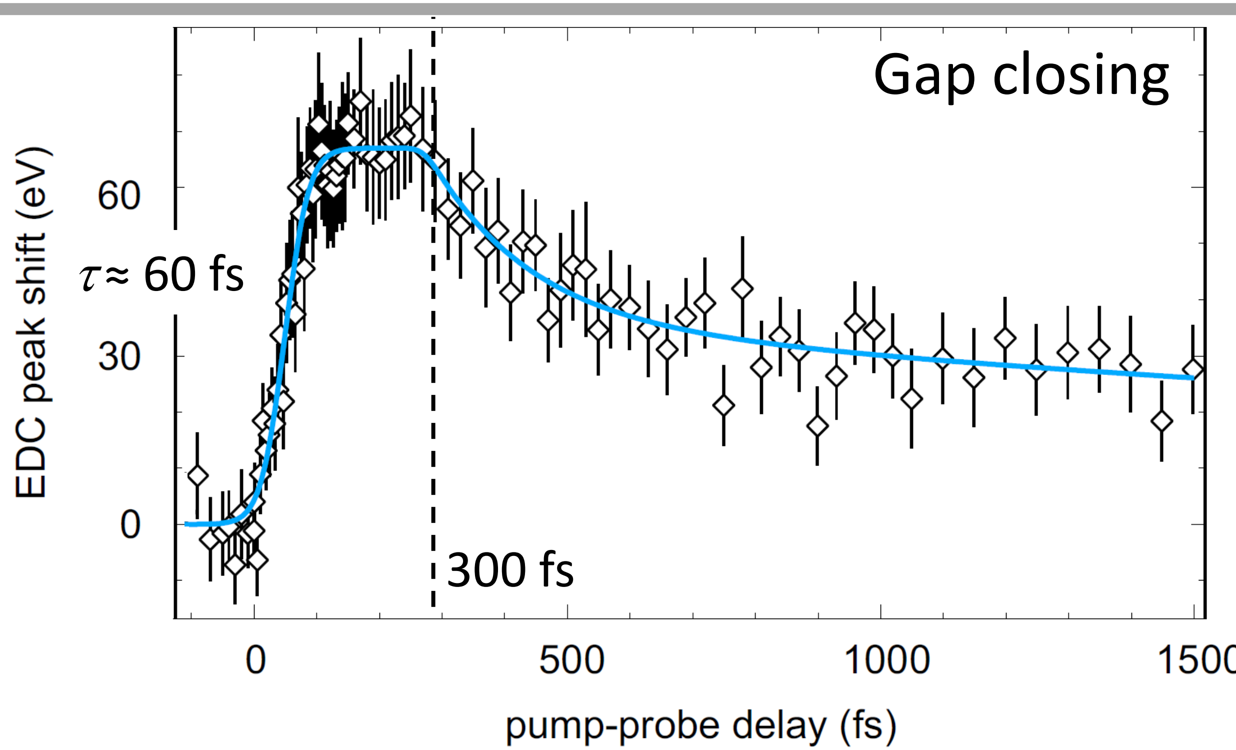
time-resolved ARPES: fluence dependence

Maximum peak-shift/temperature rise



*Perfetti *et al.*, PRB **66**, 075107 (2002)

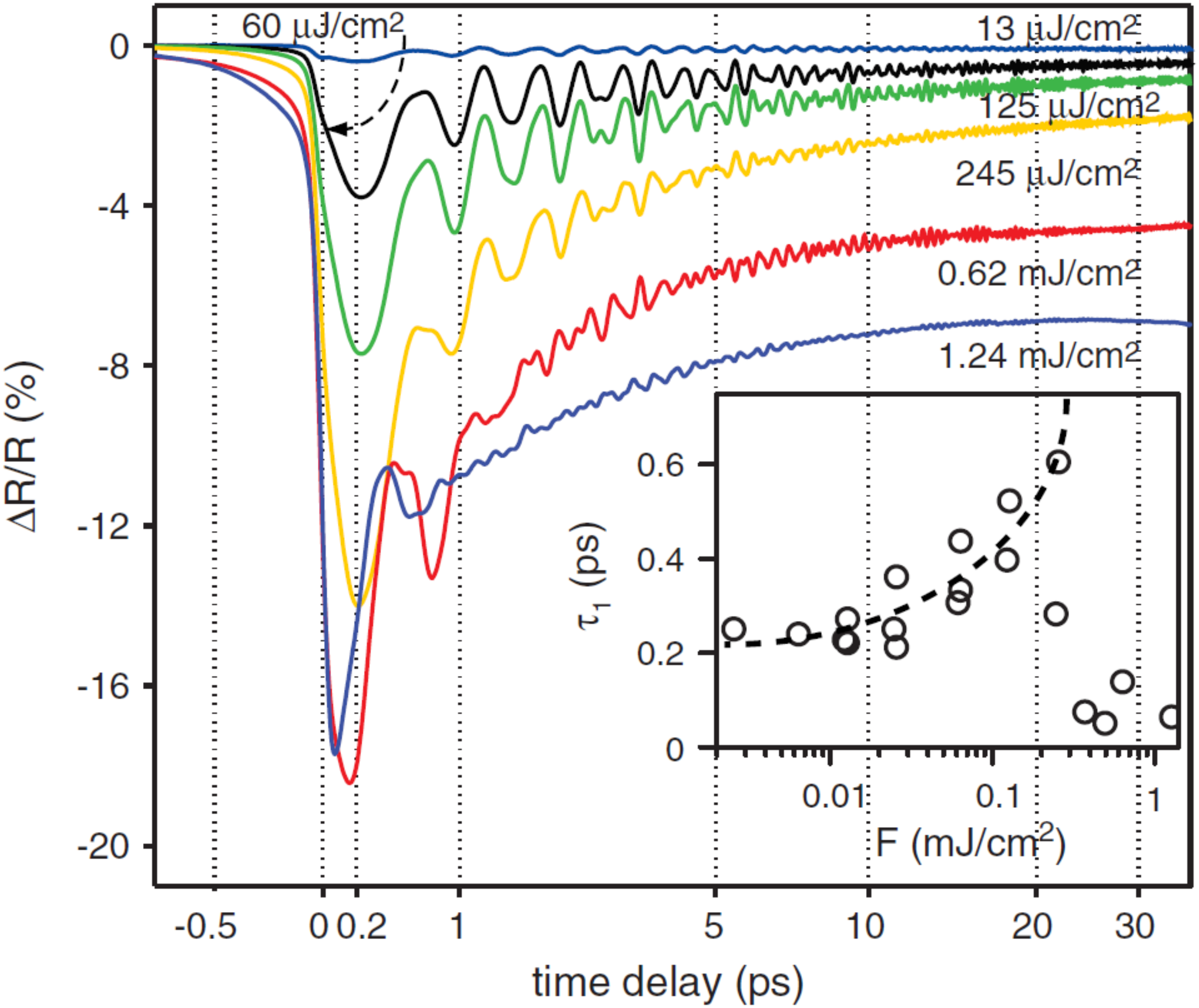
gap quenching dynamics



F_{abs} = 0.72 mJ/cm²

Prototypical Peierls insulator – A_xMoO_3

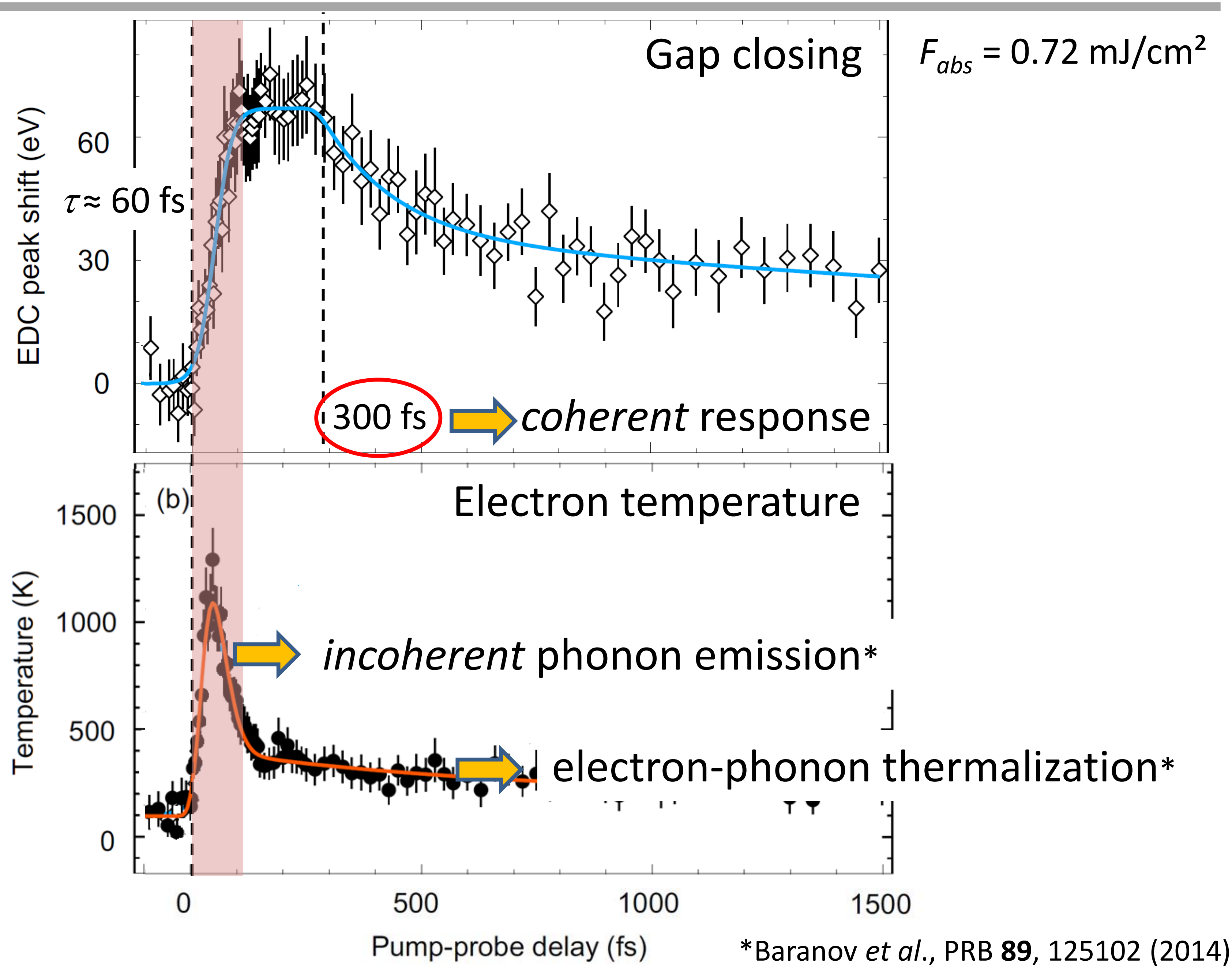
$K_{0.3}MoO_3$



(strongest) AM mode $T_{AM} \approx 600$ fs

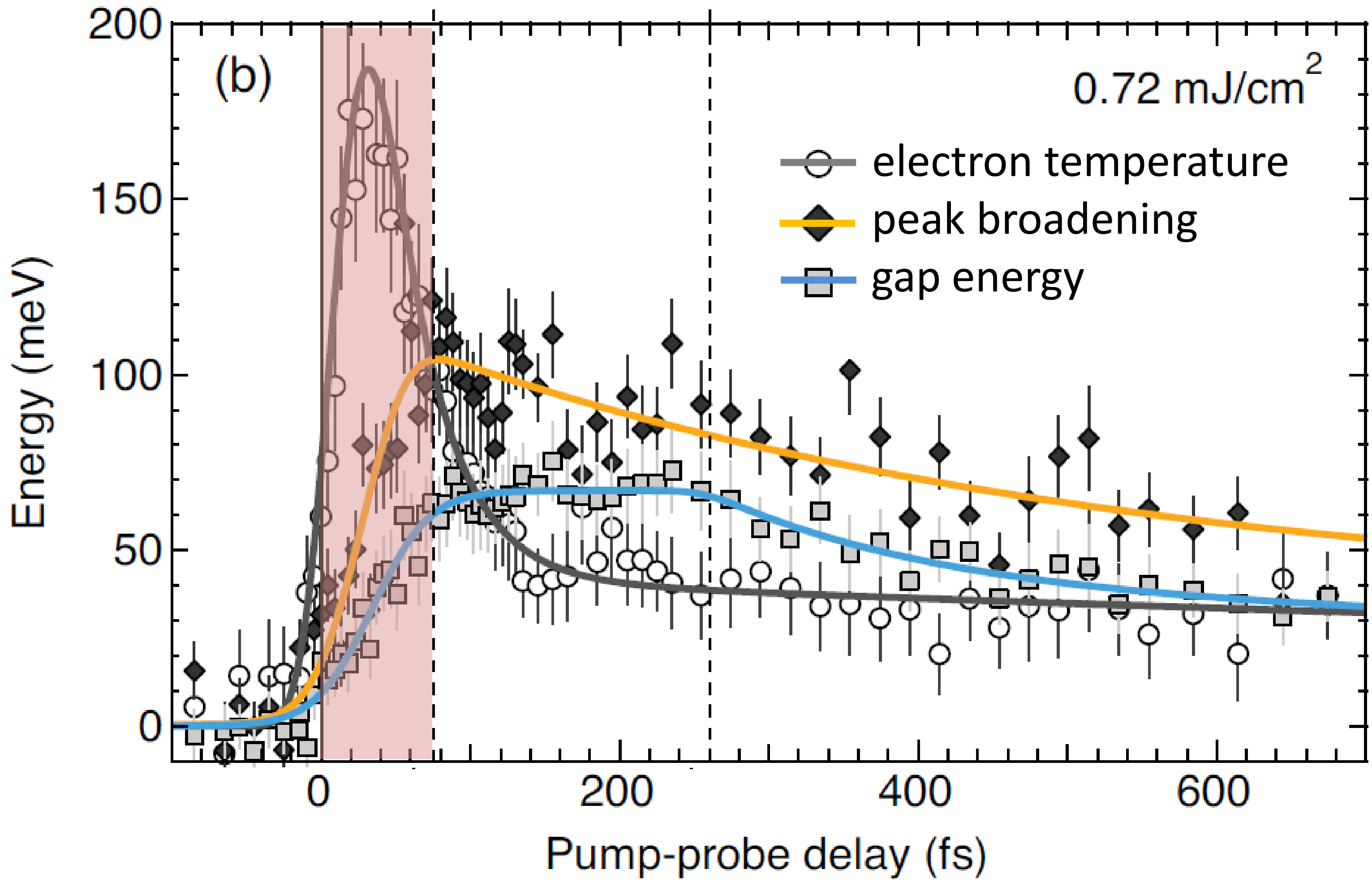
Tomeljak *et al.*, PRL **102**, 066404 (2009)

gap quenching dynamics



*Baranov *et al.*, PRB **89**, 125102 (2014)

gap quenching dynamics



Electron-phonon coupling of phason modes

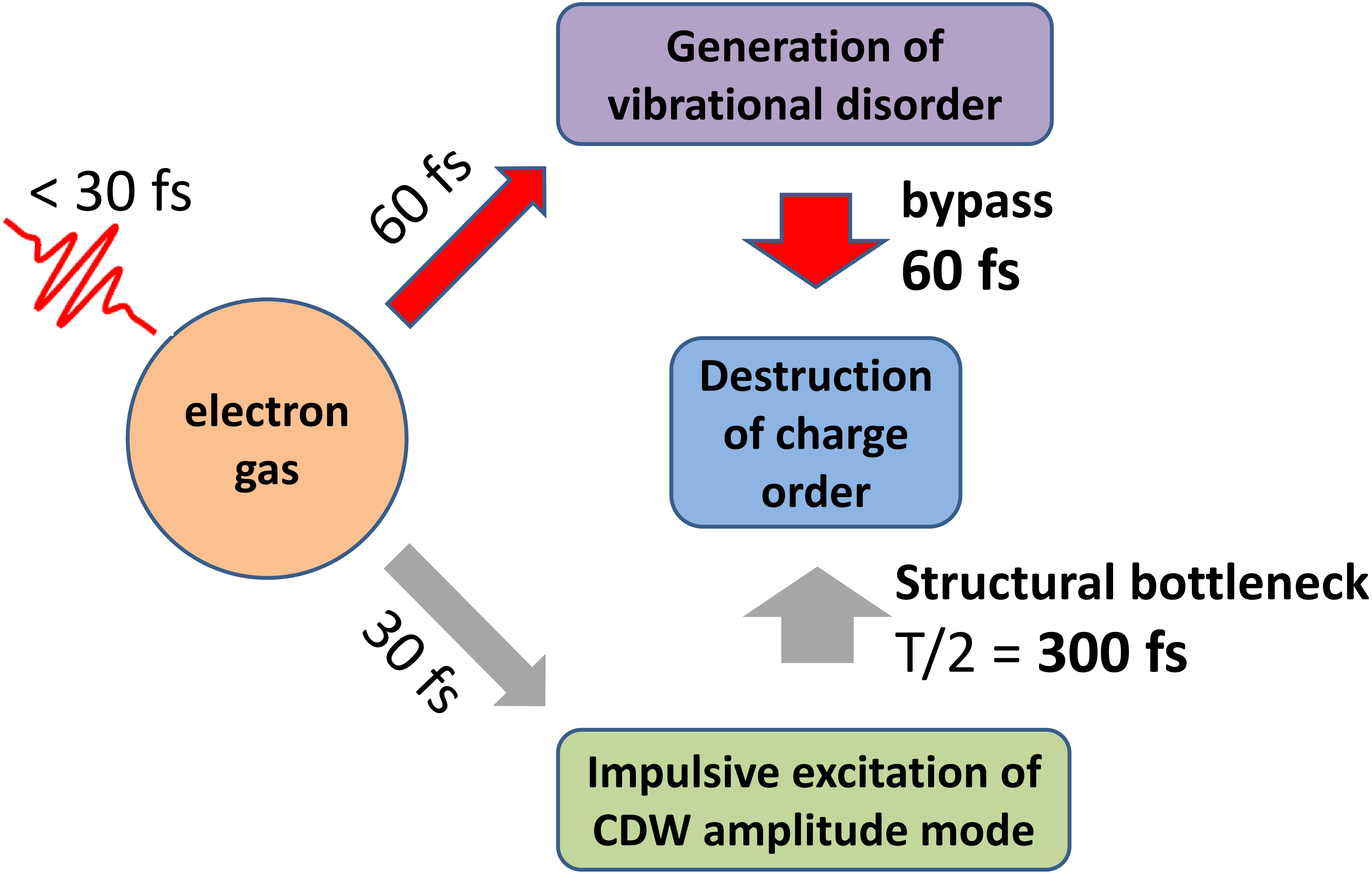
TABLE I. The dimensionless electron-phonon coupling λ_n, the frequency ω_n, and the damping Γ_n of the phase phonon modes calculated after the model of Ref. 17. A plasma frequency of $\nu_p = 2.1 \times 10^4$ cm^{-1} and a static dielectric function $\varepsilon_0 = 7 \times 10^4$ have been used (Refs. 4 and 17).

ω_n (cm^{-1})	λ_n	Γ_n (cm^{-1})	ω_n (cm^{-1})	λ_n	Γ_n (cm^{-1})
30	0.06	0.8	300	0.01	20
50	0.06	5.0	350	0.01	20
80	0.04	9.0	400	0.02	60
100	0.04	3.0	450	0.02	40
150	0.09	15	500	0.08	60
200	0.04	10	550	0.08	60
250	0.01	20	800	0.10	95

> 15 THz

L. Degiorgi, et al.,
Phys. Rev. B RC 49, 14754 (1994)

Summary



< 30 fs
electron gas
60 fs
Generation of vibrational disorder
bypass
60 fs
Destruction of charge order
30 fs
Structural bottleneck
T/2 = 300 fs
Impulsive excitation of CDW amplitude mode

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M. Aeschlimann, Univ. of Kaiserslautern
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DFG Deutsche Forschungsgemeinschaft

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