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Local and Non-Local Relaxation Dynamics of Hot Electrons in Au/Fe/MgO(001) thin films

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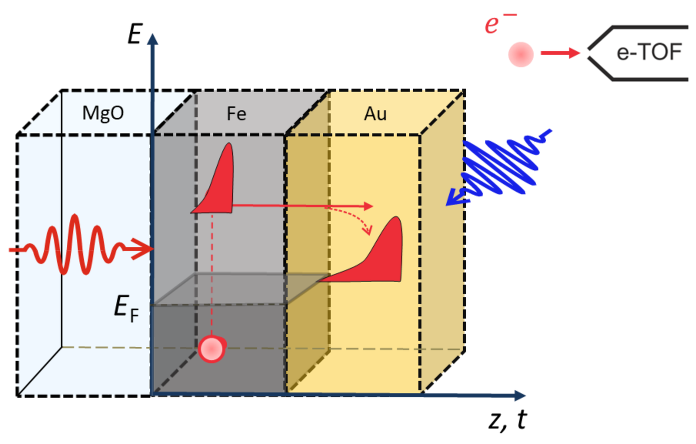
Abstract

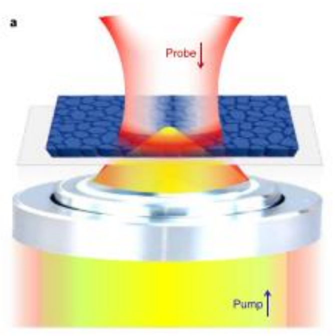
Employing femtosecond laser pulses in front and back side pumping of Au/Fe/MgO(001) combined with detection in two-photon photoelectron emission spectroscopy, we analyze local relaxation dynamics of excited electrons in buried Fe, injection into Au across the Fe-Au interface, and electron transport across the Au layer at 0.6 to 2.0 eV above the Fermi energy. By analysis as a function of Au film thickness we obtain the electron lifetimes of bulk Au and Fe and distinguish the relaxation in the heterostructures constituents. We also show that the excited electrons propagate through Au in a superdiffusive regime and conclude further that electron injection across the epitaxial interface proceeds ballistically by electron wave packet propagation.

Electron dynamics in buried media as a function of energy

By using a ***back-pump/ front-probe*** tr-photoemission technique the photoexcited hot electrons exhibit local and non-local dynamics, which leads to ***relaxation and transport*** processes.

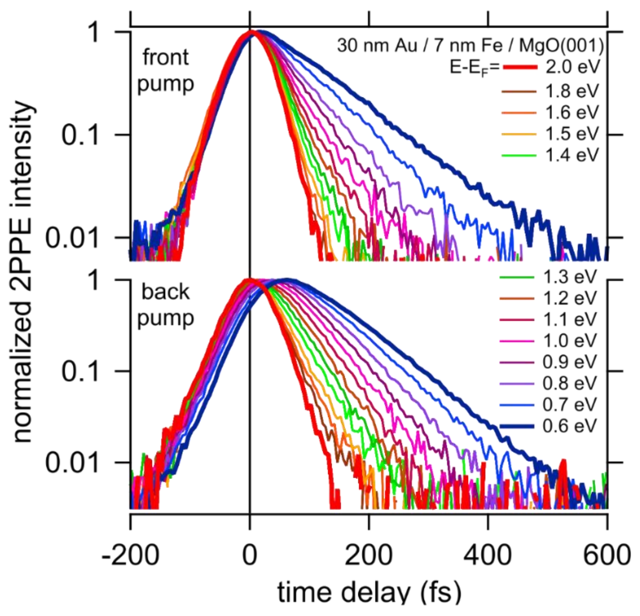
- **interplay among relaxation and transport processes of hot electrons**
  — energy E, space z and time t
  — dynamics in buried media and interfaces

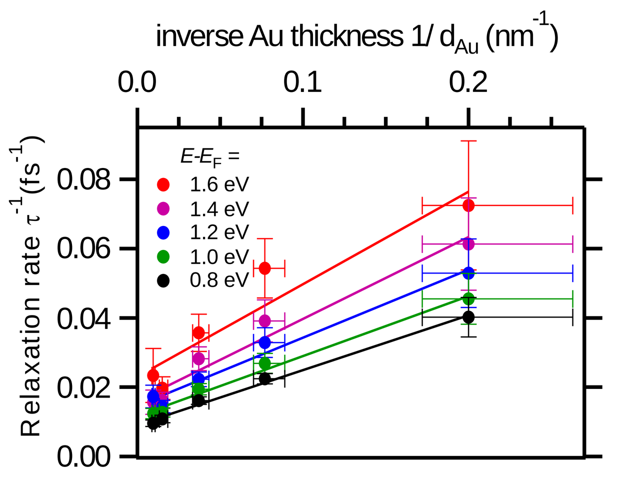




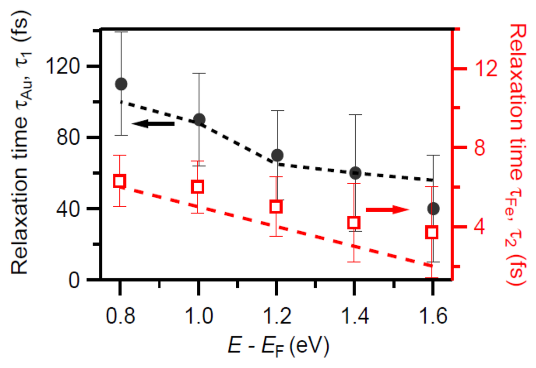
- fs-TAM on $MAPI_{3-x}Cl_x$ perovskite
  — analysis of spatiotemporal dynamics of carriers
  — missing energy *E* information

J. Sung et al., Nature Phys. ***16****, 171 (2020)*





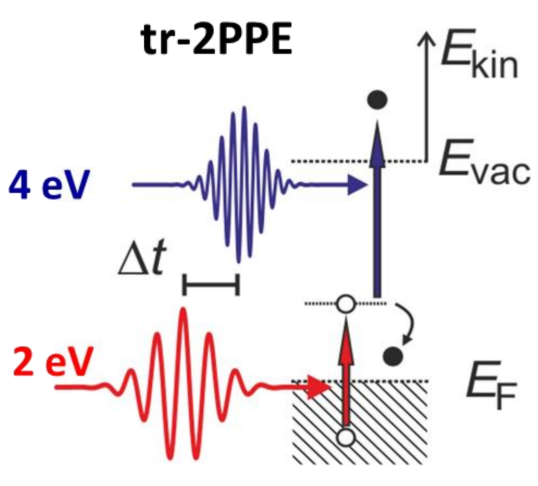
$$\tau^{-1}(d_{\mathrm{Au}}) = \tau_{\mathrm{Au}}^{-1} + \tau_{\mathrm{Fe}}^{-1} = A + \frac{B}{d_{\mathrm{Au}}}$$

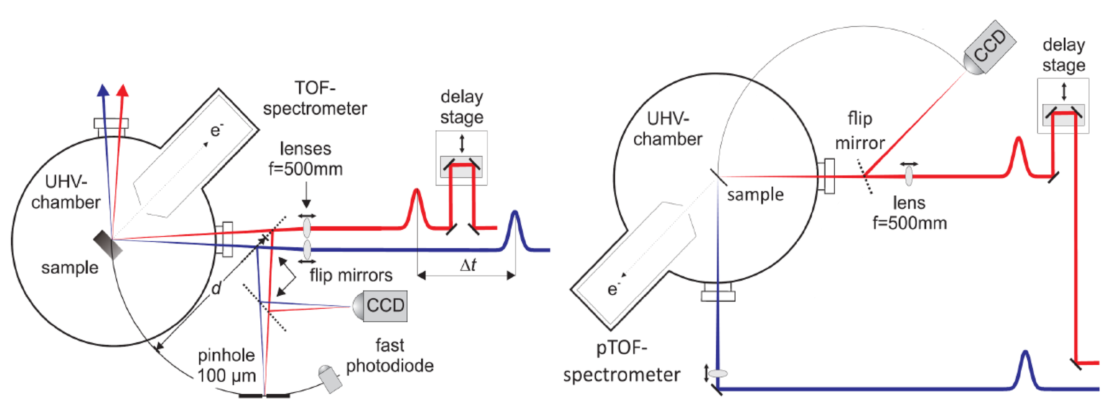


- Via back-pump data analysis:
  - superdiffusive transport behaviour of excited e⁻
  - distinguished e⁻ dynamics in the individual heterostructure constituents

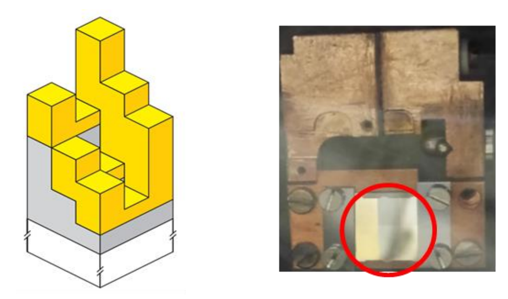
Y. Beyazit et al., PRL ***125****, 076803 (2020)*

Au/Fe/MgO(001) thin films & tr-two-photon-photoemission

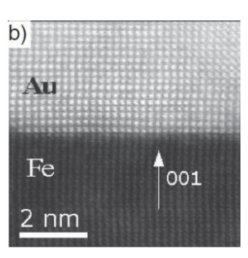




(a) Front pump front probe (b) Back pump front probe



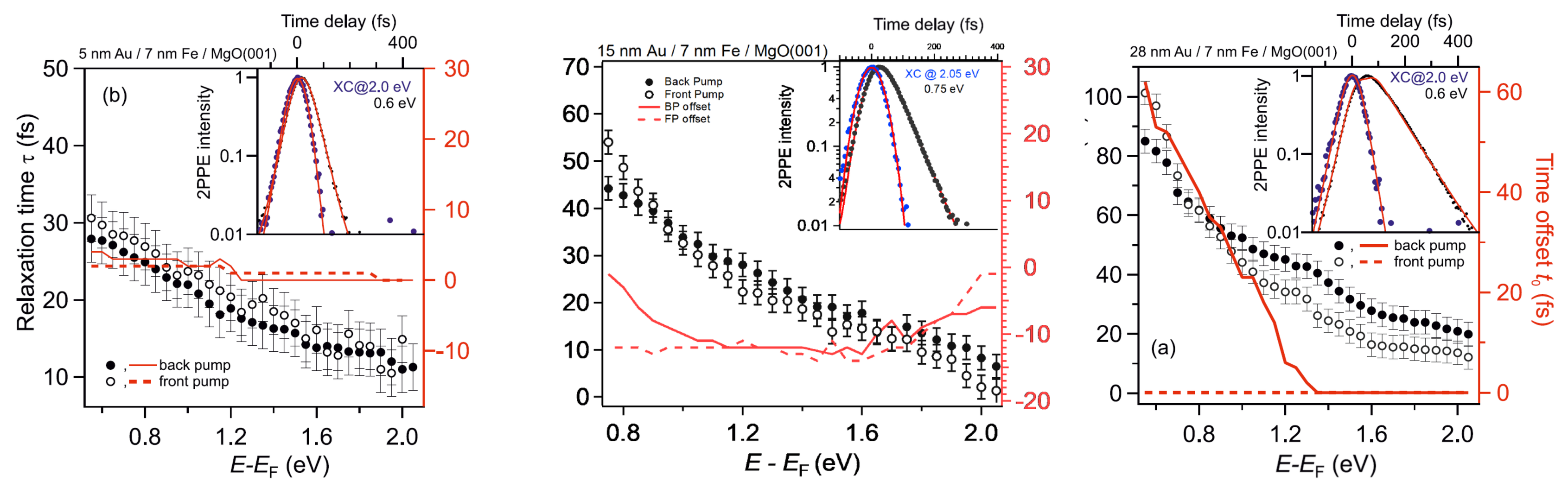
5-30 nm Au / 7-30 nm Fe / 0.5 mm MgO(001)



A. Menikov et al., PRL ***107****, 076601 (2011)*

- epitaxially grown Fe-Au layer onto each other with different thickness combination
- femtosecond time-resolved two-photon-photoemission (tr-2PPE) is employed in a pump-probe scheme; sample excited by a **2 eV** pulse with an incident ***weak*** excitation fluence *F* = 50 µJ/cm² and subsequently probed by a time delayed **4 eV** pulse. Photoemitted electrons were detected by a time-of-flight spectrometer (e-TOF)
- after finding spatial and temporal overlap of both beams on the sample, **time-zero** is determined by a Gaussian fit at the highest energy population curve → **cross-correlation** XC = 75 fs
- compare obtained spectra for different film thickness to observe electron transport

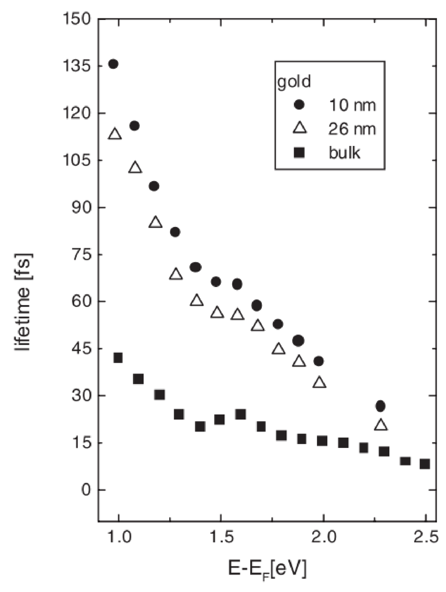
Relaxation dynamics τ_{FP} vs. τ_{BP}

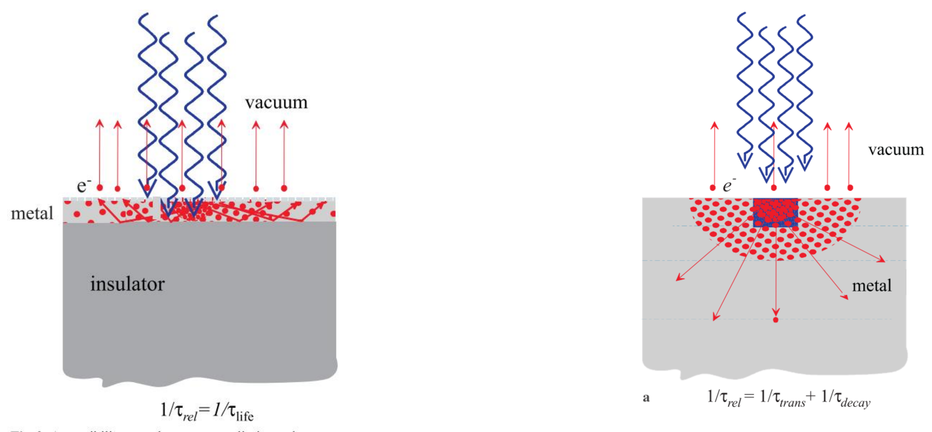


- $\tau_{FP}(E-E_F) \approx \tau_{BP}(E-E_F)$
- $E - E_F = 1-2\ eV$: $\tau_{FP} \leq \tau_{BP}$
- $E - E_F = 0.75-1\ eV$: $\tau_{FP} \geq \tau_{BP}$
- $E - E_F = 1-2\ eV$: $\tau_{FP} < \tau_{BP}$
- $E - E_F = 0.60-1\ eV$: $\tau_{FP} > \tau_{BP}$

➢ τ_{FP} vs. τ_{BP} thickness- and energy-dependent, why?

Optical penetration and transport effects

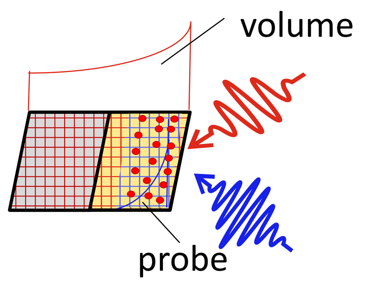




M. Aeschlimann et al., Appl. Phys. ***A 71****, 485–491 (2000)*

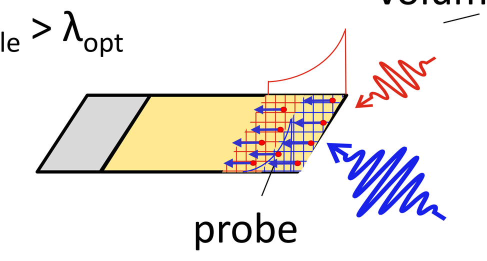
- τ decreases with increased sample thickness d
- population decay of hot e's near to surface due to transport
- d_{sample} vs. λ_{opt} important

thin films
$d_{sample} < \lambda_{opt}$



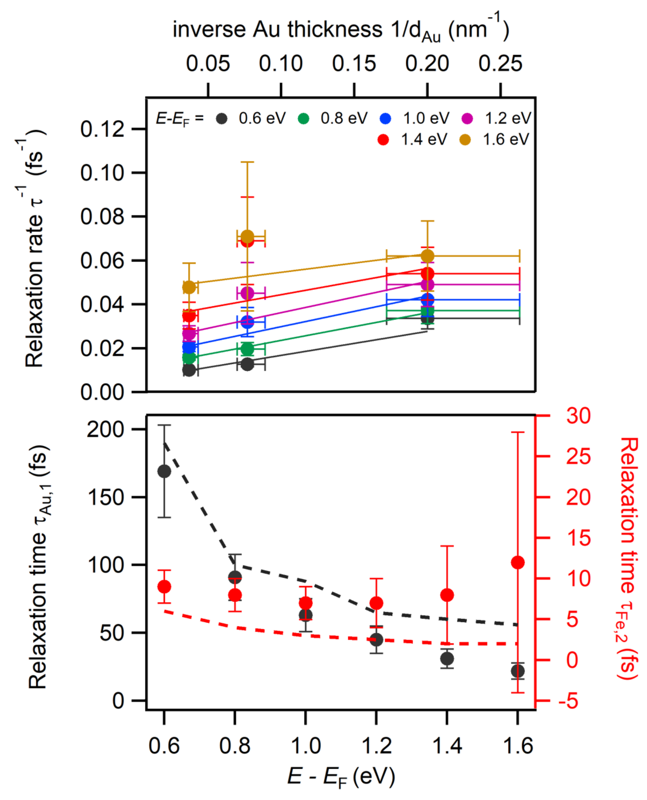
no gradient $\nabla f(E) = 0$ ➜ no transport

thick films
$d_{sample} > \lambda_{opt}$



gradient $\nabla f(E) \neq 0$ ➜ transport

obtained with FP data



- front side pumping:
  - population decay of hot e's near Au surface region due to transport into bulk
  - Fe layer as sink; additional decay channel
- seperation of e⁻ dynamics in individual constituents of the heterostructure **not clear**

Outlook

- **FP** measurements with **thicker** d_{Au}
- wedge sample: analysis of ballistically propagating e's