Quantum quench of Kondo correlations in optical absorption

Initial question: Can Kondo effect be detected by purely optical studies?

Rolf Helmes, Michael Sindel, Laszlo Borda, Jan von Delft (LMU)

First proposal: PRB, **72**, 125301 (2005)

Hakan Tureci (Princeton), Martin Claassen, Atac Imamoglu (ETH),
Markus Hanl, Andreas Weichselbaum, Theresa Hecht, Jan von Delft (LMU)
Bernd Braunecker (Basel), Sasha Govorov (Ohio), Leonid Glazman (Yale)

Full theory: PRL, **106**, 107402 (2011)

Christian Latta, F. Haupt, P. Fallahi, S. Faelt, Hakan Tureci, Atac Imamoglu (ETH),
Markus Hanl, Andreas Weichselbaum, Jan von Delft (LMU), Leonid Glazman (Yale)

Experiment: Nature, **474**, 627 (2011)

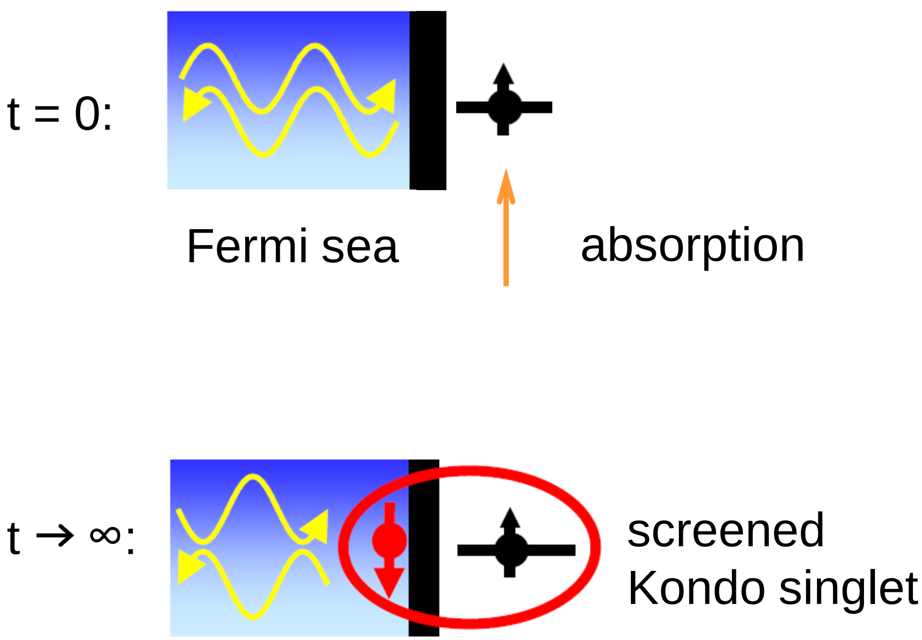
Final punchline: Local quantum quench induces tunable Anderson orthogonality catastrophe, directly observed in optical absorption lineshape!

LMU | Arnold Sommerfeld Center for Theoretical Physics | CeNS Center for NanoScience | LMU | nim nanosystems initiative munich | SFB/TR-12 | SFB 631

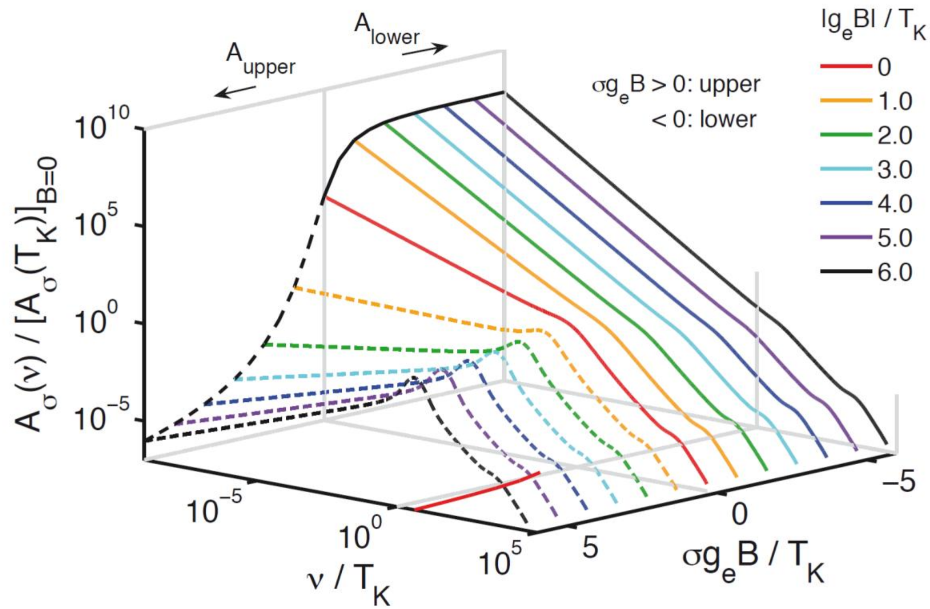
Quantum quench of Kondo correlations in optical absorption [theory]

Hakan Tureci (Princeton), Martin Claassen, Atac Imamoglu (ETH),
Markus Hanl, Andreas Weichselbaum, Theresa Hecht, Jan von Delft (LMU)
Bernd Braunecker (Basel), Sasha Govorov (Ohio), Leonid Glazman (Yale)

[PRL 2011]



tunable orthogonality exponents predicted

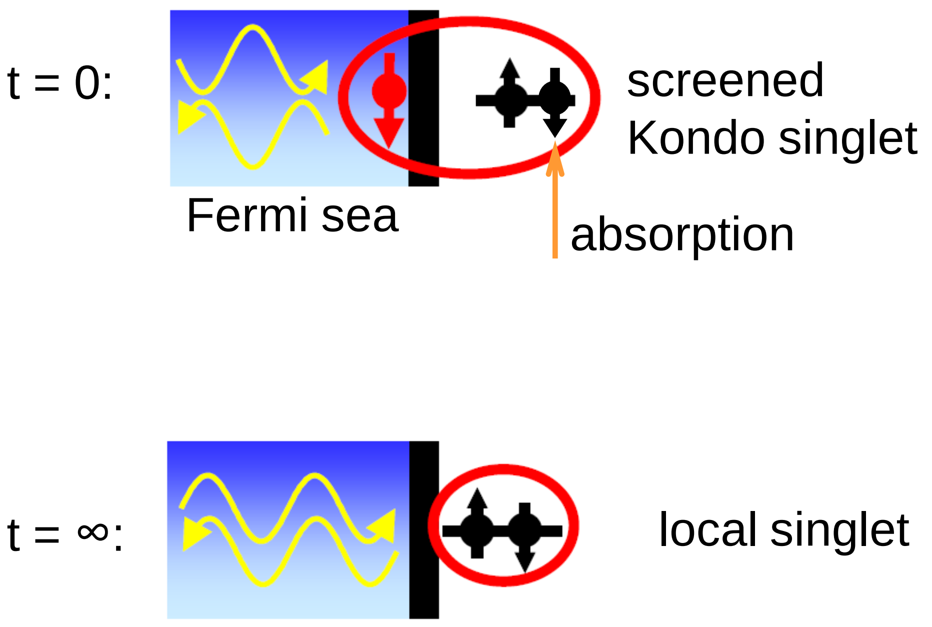


What happens when an optical excitation is used to “switch on” Kondo correlations?

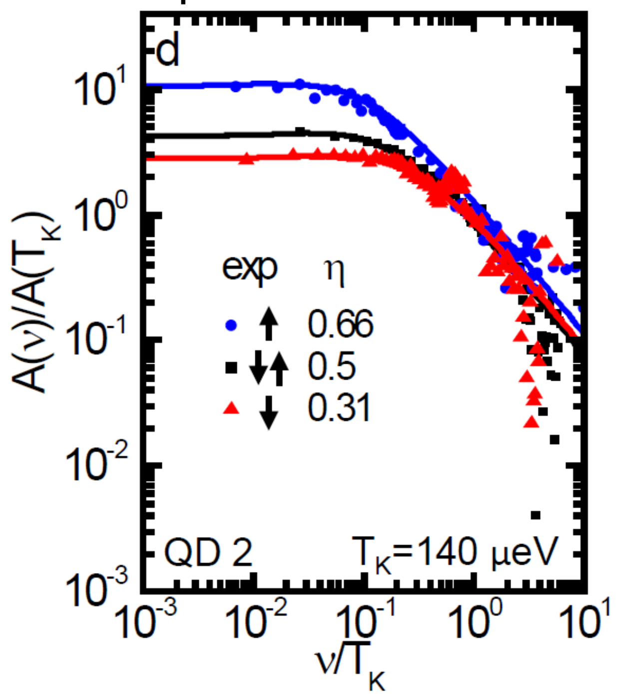
Quantum quench of Kondo correlations in optical absorption [experiment]

Christian Latta, F. Haupt, P. Fallahi, S. Faelt, Hakan Tureci, Atac Imamoglu (ETH), Markus Hanl, Andreas Weichselbaum, Jan von Delft (LMU), Leonid Glazman (Yale)

[arXiv:1102.3982v1]



tunable orthogonality exponents observed



What happens when an optical excitation is used to “switch off” Kondo correlations?

Outline

Reminder: Kondo effect in transport

Proposed experimental setup

Theoretical predictions for lineshape:

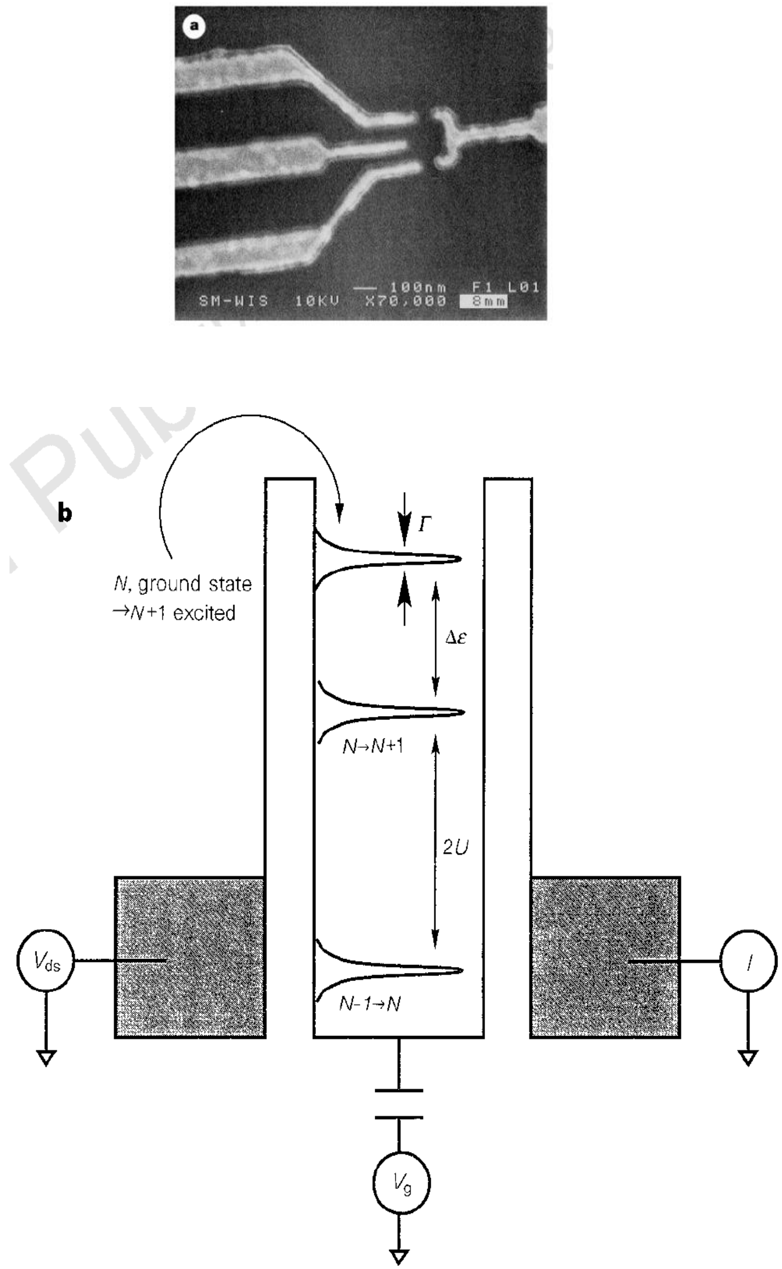
- scaling
- Anderson orthogonality power laws
- magnetic field

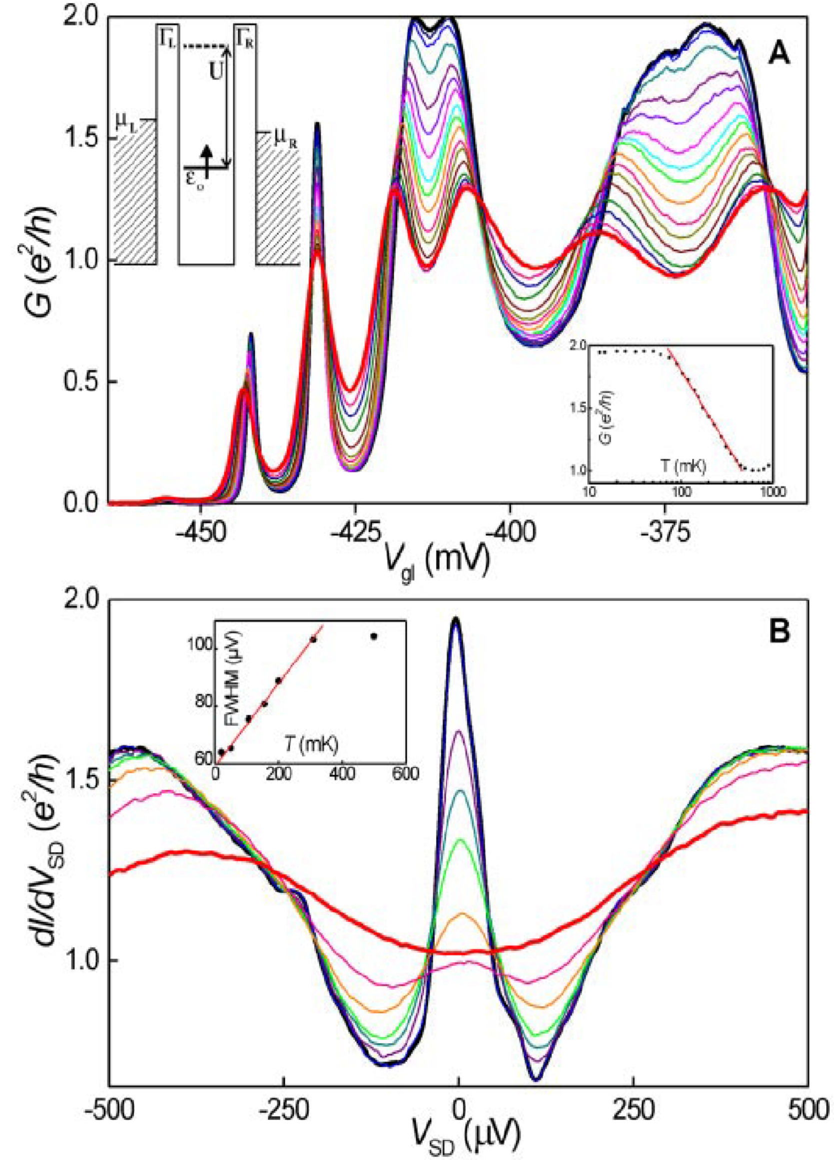
Experimental realization and results

Outlook

Kondo effect in transport

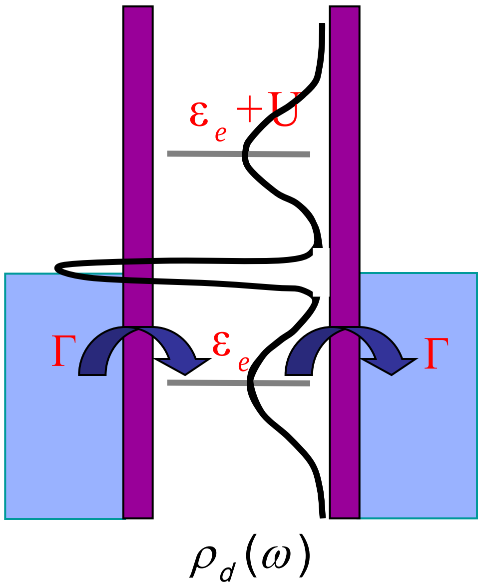
Goldhaber-Gordon et al., Nature **391**, 156 (1998)
Cronenwett et al., Science **281**, 540 (1998)
Simmel et al., PRL **83**, 804 (1999)





Anderson model

P.W. Anderson (1961)

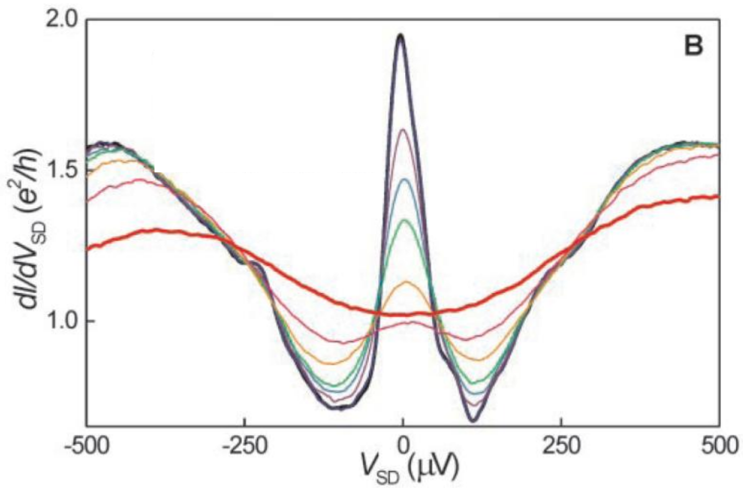


parameters (all tunable!):

local energy level $\varepsilon_e \propto V_g$

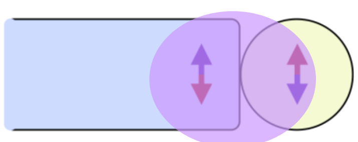
charging energy U

level width Γ



$$H = \sum_{k,\sigma} \varepsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} + \sum_{\sigma} \varepsilon_e d_\sigma^\dagger d_\sigma + U n_{e\downarrow} n_{e\uparrow} + \sum_{k,\sigma} V_k (c_{k\sigma}^\dagger e_\sigma + e_\sigma^\dagger c_{k\sigma})$$

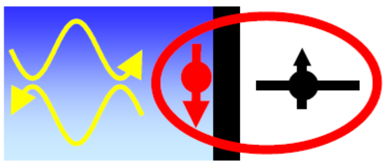
new (tunable!) low-energy scale: $T_K = \frac{1}{2}\sqrt{U\Gamma}\, e^{\pi\varepsilon_e(\varepsilon_e+U)/\Gamma U}$



For $T<T_K$, local spin screened into singlet: "Kondo effect"

local density of states develops "Kondo resonance"

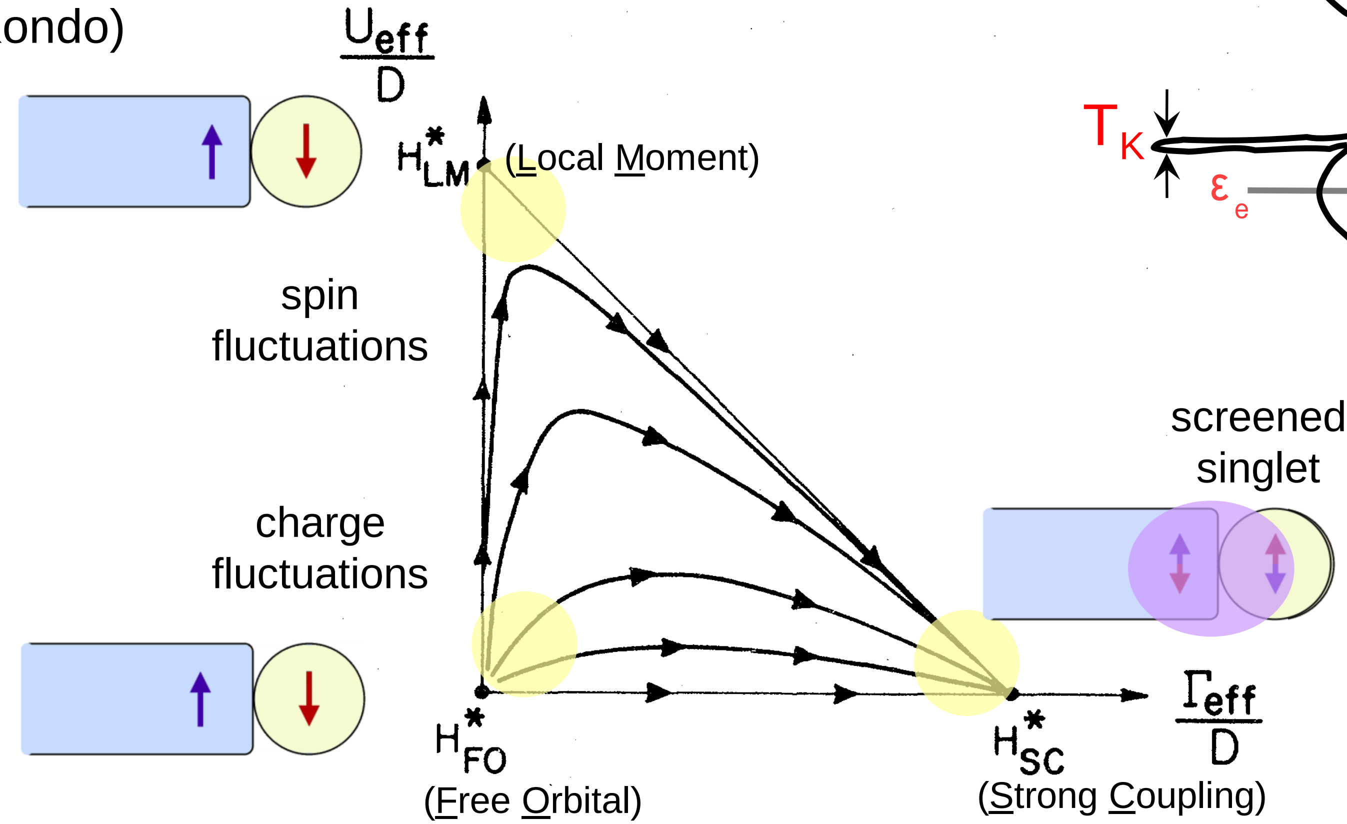
other electrons experience strong phase shift

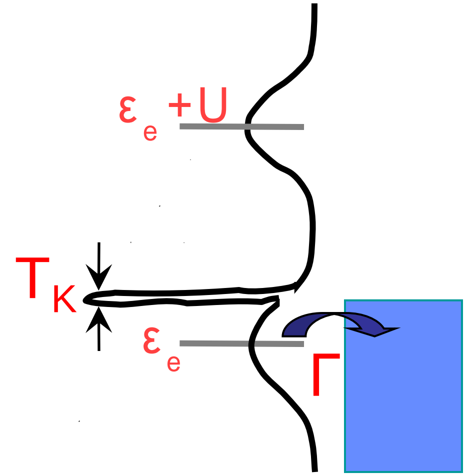


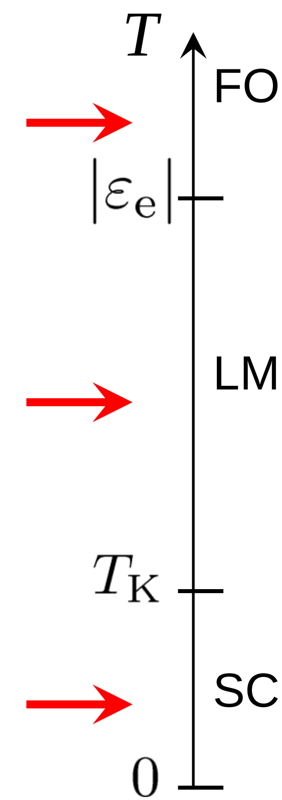
Anderson Model

$$H_{\mathrm{LM}} = \sum_{k\sigma} \varepsilon_{k\sigma} c^{\dagger}_{k\sigma} c_{k\sigma} + \frac{J}{\rho} \vec{s}_{\mathrm{e}} \cdot \vec{s}_{\mathrm{c}}$$

(Kondo)







$$H_{\mathrm{FO}} = \sum_{k\sigma} \varepsilon_{k\sigma} c^{\dagger}_{k\sigma} c_{k\sigma}$$

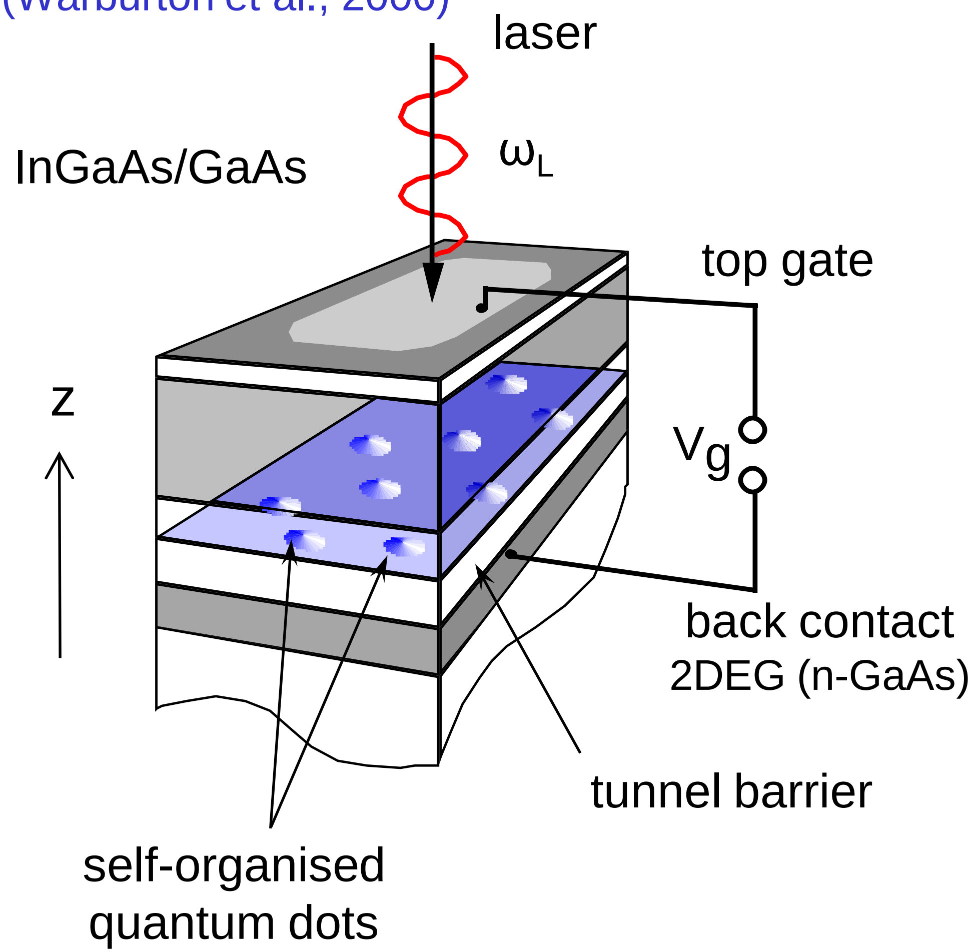
$$+ \sum_{\sigma} \varepsilon_{\mathrm{e}\sigma} n_{\mathrm{e}\sigma} + U n_{\mathrm{e}\uparrow} n_{\mathrm{e}\downarrow} + \sqrt{\frac{\Gamma}{\pi\rho}} \sum_{\sigma} (e^{\dagger}_{\sigma} c_{k\sigma} + \mathrm{h.c.})$$

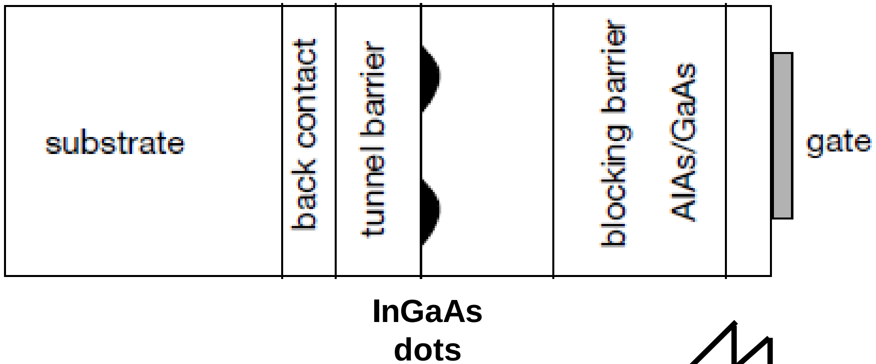
(Anderson)

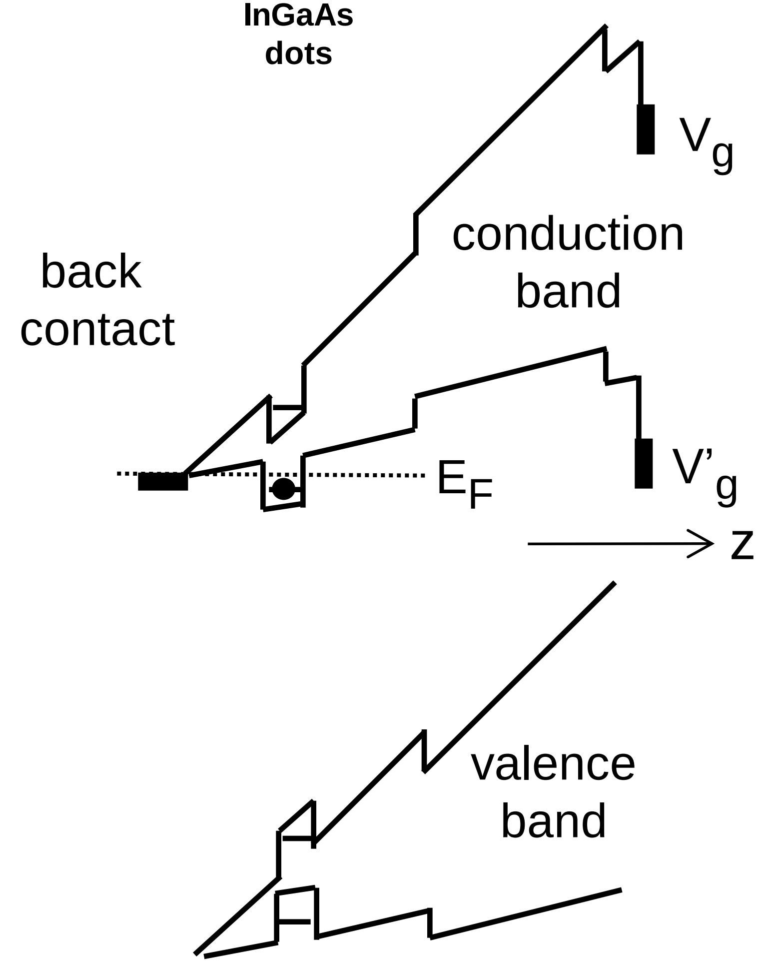
$$T_{\mathrm{K}} = \sqrt{\frac{\Gamma U}{2}}\, e^{-\frac{\pi |\varepsilon^{\mathrm{f}}_{\mathrm{e}}(\varepsilon^{\mathrm{f}}_{\mathrm{e}}+U)|}{(2U\Gamma)}}$$

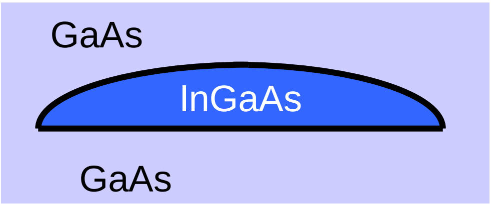
Experimental Setup

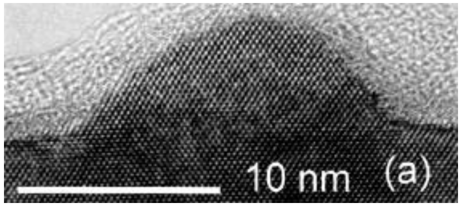
(Warburton et al., 2000)





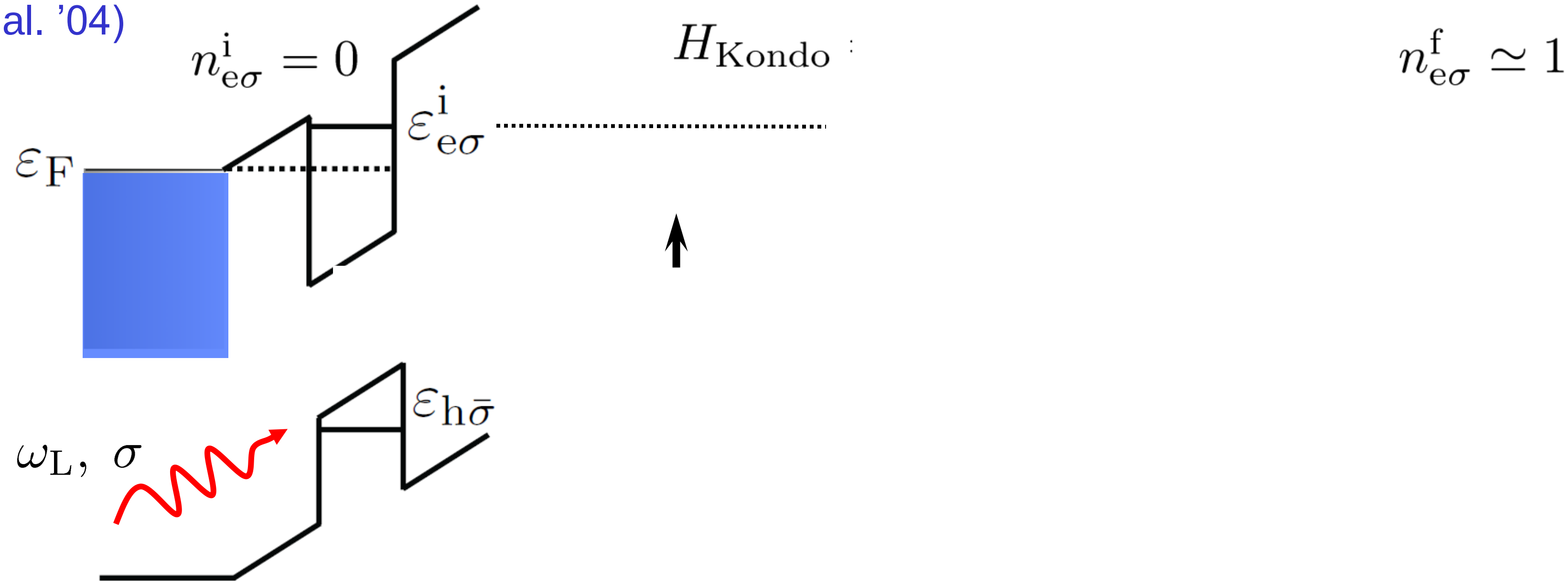






Proposed Experiment: Absorption in X^0 transition

(Helmes et al. '04)



initial

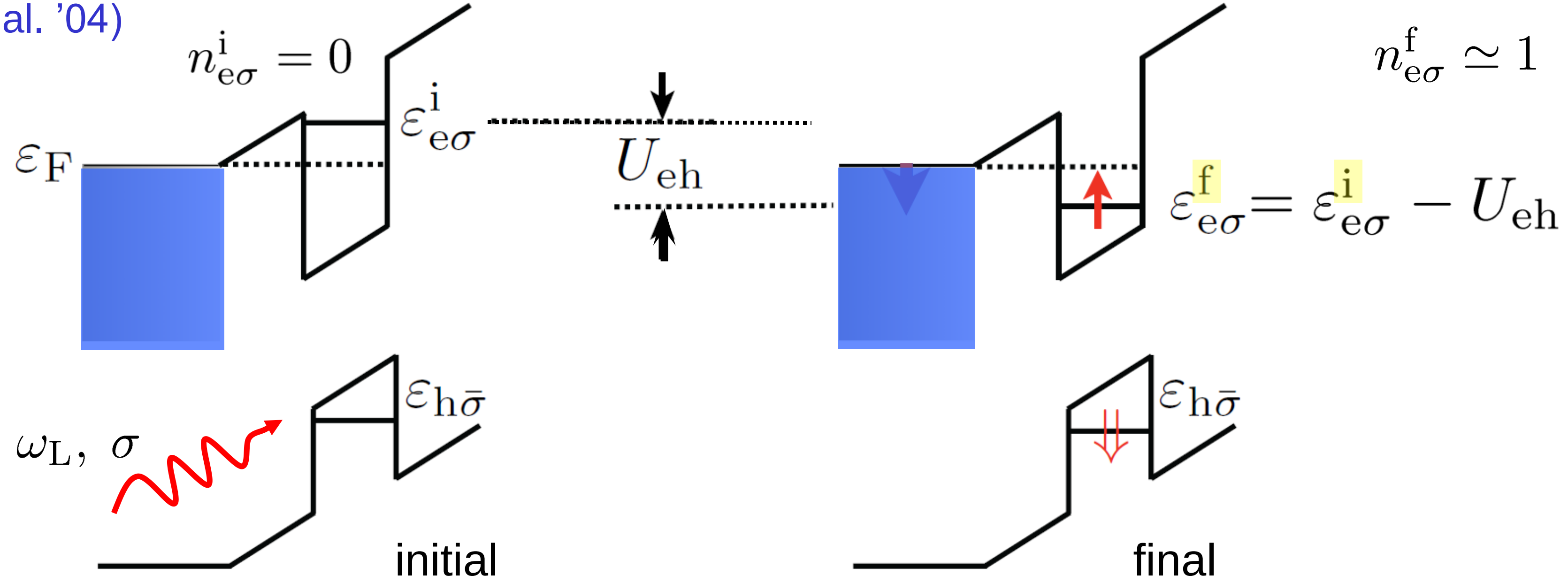
Optical absorption induces a quantum quench: $H^{\mathrm{initial}} \neq H^{\mathrm{final}}$

What is subsequent transient dynamics of dot + Fermi-sea ?

Transient dynamics after Kondo interaction is suddenly switched on ?

Hamiltonian

(Helmes et al. '04)



Anderson model (AM)

$$H^{\mathrm{i/f}} = H^{\mathrm{i/f}}_{\mathrm{QD}} + \sum_{k\sigma} \varepsilon_{k\sigma} c^{\dagger}_{k\sigma} c_{k\sigma} + \sqrt{\Gamma/\pi\rho} \sum_{\sigma} (e^{\dagger}_{\sigma} c_{\sigma} + \mathrm{h.c.})$$

$$H^{\mathrm{i}}_{\mathrm{QD}} = \sum_{\sigma} \varepsilon^{\mathrm{i}}_{\mathrm{e}\sigma} n_{\mathrm{e}\sigma} + U n_{\mathrm{e}\uparrow} n_{\mathrm{e}\downarrow}$$

$$c_{\sigma} = \sum_{k} c_{k\sigma} = \psi_{\sigma}(0)$$

$$H^{\mathrm{f}}_{\mathrm{QD}} = \sum_{\sigma} \varepsilon^{\mathrm{f}}_{\mathrm{e}\sigma} n_{\mathrm{e}\sigma} + U n_{\mathrm{e}\uparrow} n_{\mathrm{e}\downarrow} + \varepsilon_{\mathrm{h}\bar{\sigma}}$$

SAM: $\varepsilon^{\mathrm{f}}_{\mathrm{e}\sigma} = -U/2;\ n^{\mathrm{f}}_{\mathrm{e}\sigma} = 1$

(symmetric Anderson model)

Dynamical correlation functions with Wilson's NRG

1989: Sakai, Shimizu, Kasuya / Costi, Hewson

1990: Yosida, Whitaker, Oliveira

1994: Costi, Hewson, Zlatic — - Transport properties (resistivity)

1999: Bulla, Hewson, Pruschke — - Patching rules for combining data from seve

2000: Hofstetter — - DM-NRG (accurate ground state needed also for high-frequency information)

2004: Helmes, Sindel, Borda von Delft — - Absorption/emission spectra after quantum quench

2005: Anders & Schiller — - Complete Fock space basis for t-NRG

2005: Verstraete, Weichselbaum, Schollwöck, von Delft, Cirac — - Relation between NRG & DMRG via MPS

2007: Peters, Anders, Pruschke — - Sum-rule-conserving spectral functions (single-shell DM)

2007: Weichselbaum & von Delft — - First truly "clean" algorithm for spectral functions at finite temperatures (full multi-shell DM)

2008: Weichselbaum, Verstraete Schollwöck, von Delft, Cirac — -Non-logarithmic discretization for split Kondo resonance

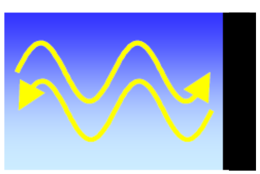
2008: Toth, Moca, Legeza, Zarand — - Flexible NRG code with non-Abelian symmetries

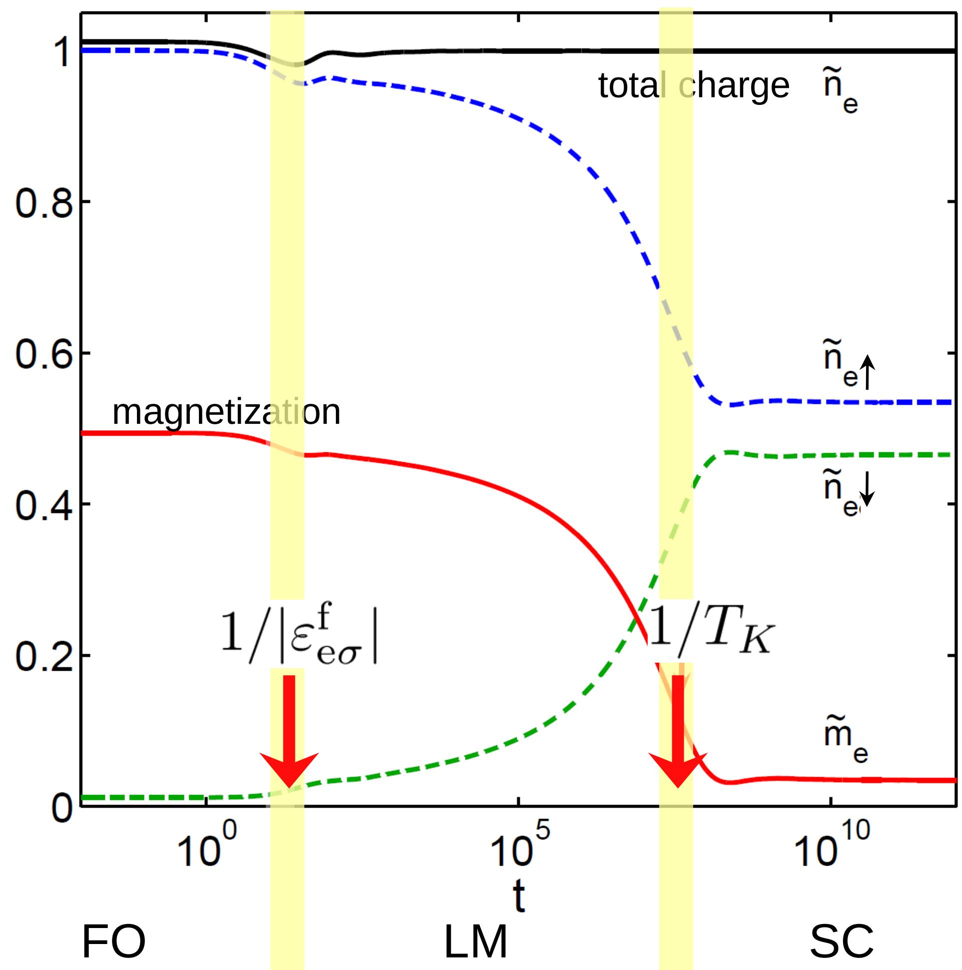
2009: Anders — - Nonequilbrium correlators via scattering state NRG

Transient Relaxation (X^0 transition)

$$\widetilde{n}_{e\sigma'}(t) = \langle \Psi_0 | e^{iH^f} \hat{n}_{e\sigma'} e^{-iH^f t} | \Psi_0 \rangle$$

t < 0

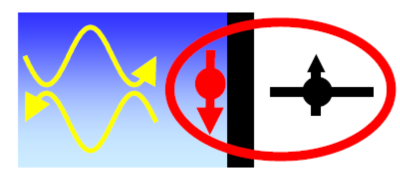


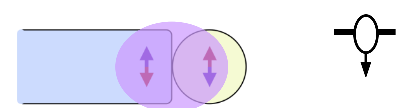


t-NRG:
Anders, Schiller '05

nonzero final magnetizaton is finite-size effect

t = ∞





Absorption Lineshape (log-linear) [SAM]

$$A_\sigma(\nu) = 2\pi \sum_{\alpha\beta} \rho^{\mathrm{i}}_\alpha \left| {}_{\mathrm{f}}\langle \beta | e^\dagger_\sigma | \alpha \rangle_{\mathrm{i}} \right|^2 \delta(\overbrace{\nu + \omega_{\mathrm{th}}}^{\omega_L} - E^{\mathrm{f}}_\beta + E^{\mathrm{i}}_\alpha)$$

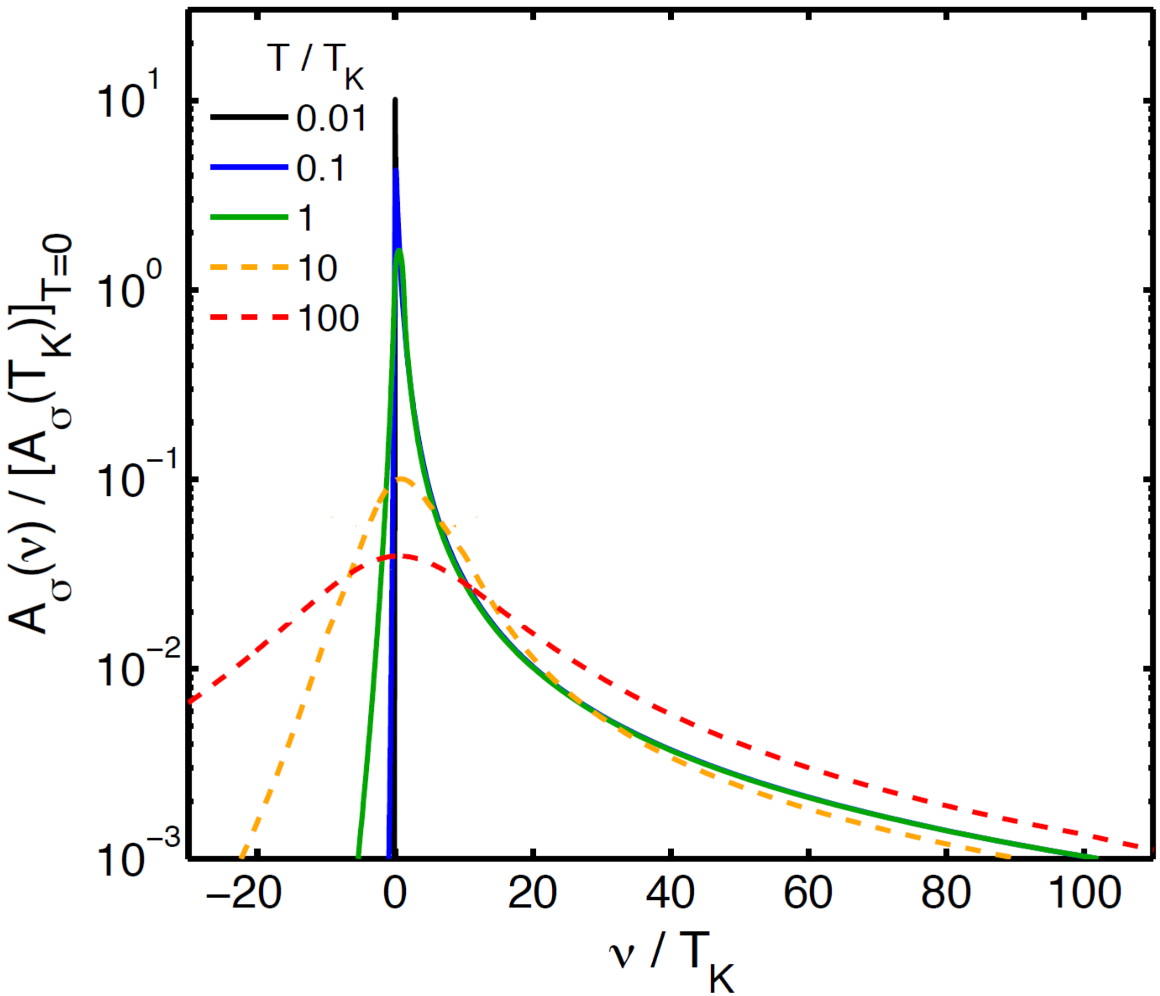
ω_L ← laser frequency

ν: detuning

ω_{th}: threshold for absorption

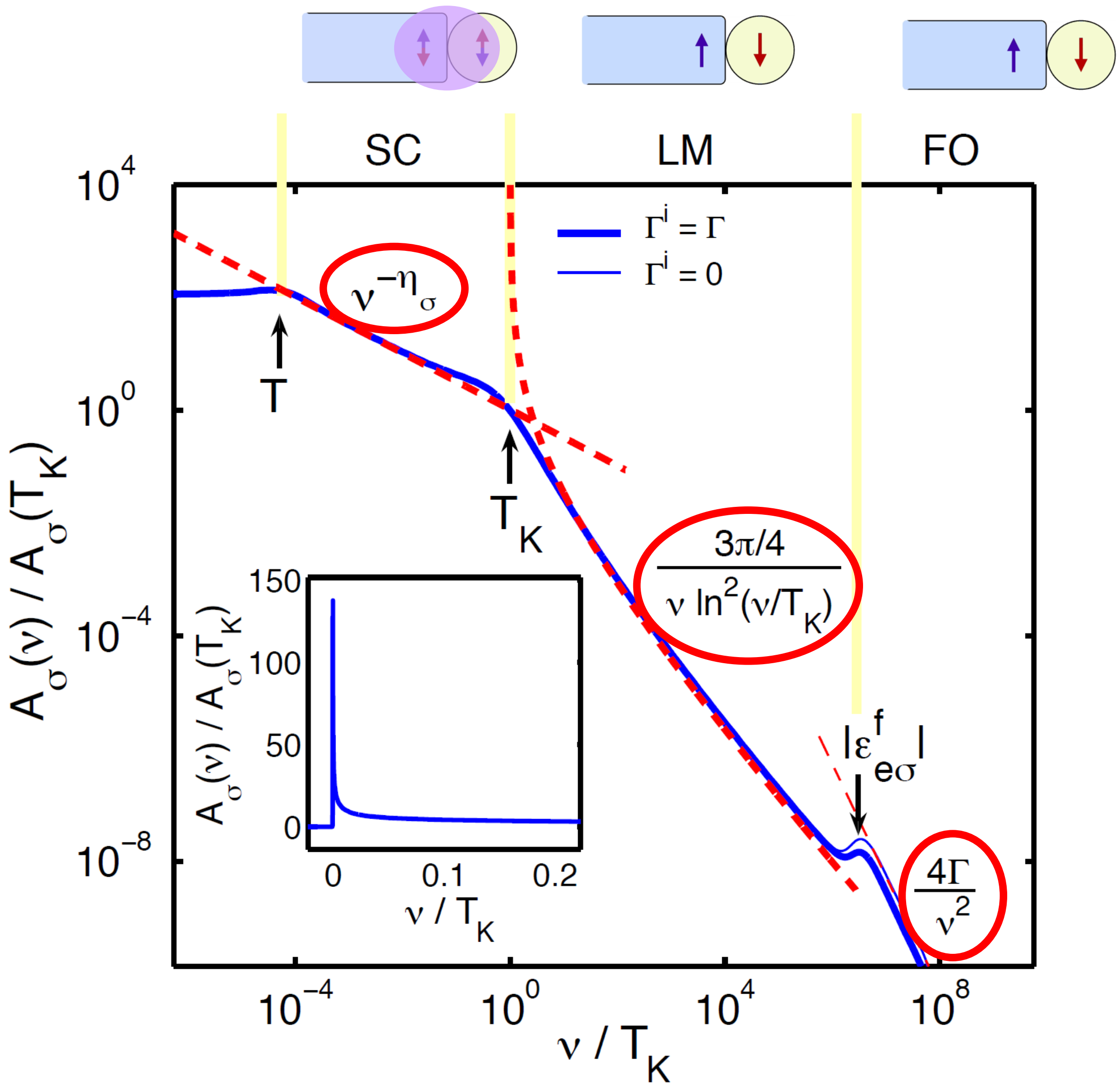
Properties of lineshape:

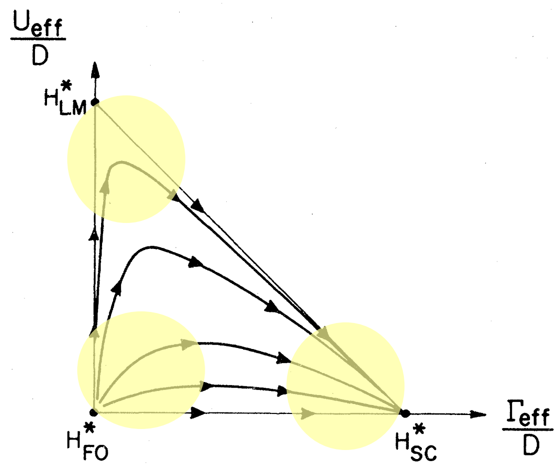
- depends on initial <u>and</u> final eigenstates
- is roughly symmetric at large T
- as T decreases, lineshape develops asymmetric threshold behavior
-and peak becomes narrower and sharper
- for T→0, lineshape shows power-law singularity

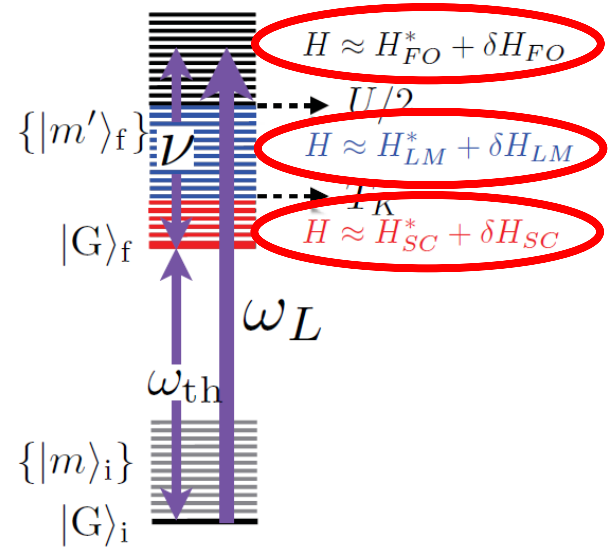


Absorption Lineshape (log-log): T = 0 [SAM]

$$A_\sigma(\nu) \;=\; 2\pi \sum_\beta \left|{}_{\rm f}\langle\beta|e^\dagger_\sigma|{\rm G}\rangle_{\rm i}\right|^2 \delta(\nu + \omega_{\rm th} - E^{\rm f}_\beta + E^{\rm i}_{\rm G})$$



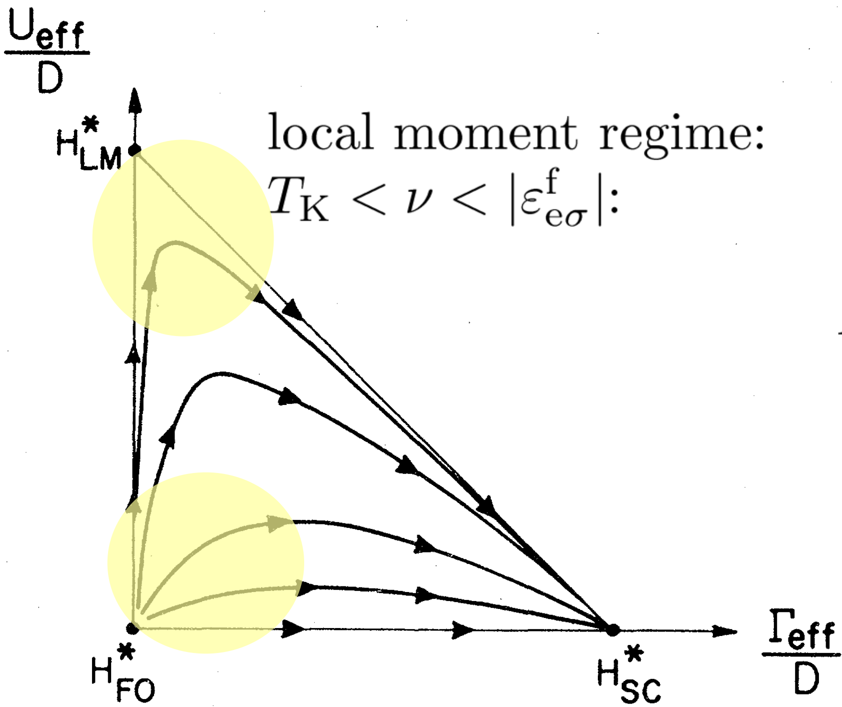




FPPT: Fixed-Point Perturbation Theory (FO, LM)

$$A_\sigma(\nu) = -2\mathrm{Im}\left[{}_{\mathrm{i}}\langle \mathrm{G}| e_\sigma \frac{1}{\nu + i0^+ - H^{\mathrm{f}} + E_{\mathrm{G}}^{\mathrm{i}}} e_\sigma^\dagger |\mathrm{G}\rangle_{\mathrm{i}} \right]$$

near fixed point: $H^{\mathrm{f}} = H^* + H'$, expand in H'



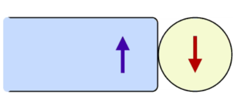
local moment regime:
$T_{\mathrm{K}} < \nu < |\varepsilon^{\mathrm{f}}_{\mathrm{e}\sigma}|$:

$$H^*_{\mathrm{LM}} = \sum_{k\sigma} \varepsilon_{k\sigma} c^\dagger_{k\sigma} c_{k\sigma} , \quad H'_{\mathrm{LM}} \frac{J}{\rho} \vec{s}_{\mathrm{e}} \cdot \vec{s}_{\mathrm{c}}$$

$$A^{\mathrm{LM}}_\sigma(\nu) = \frac{3\pi}{4} \frac{J^2(\nu)}{\nu}$$

$$J(\nu) = \frac{1}{\ln(\nu/T_{\mathrm{K}})} \quad \text{(rescaled coupling)}$$

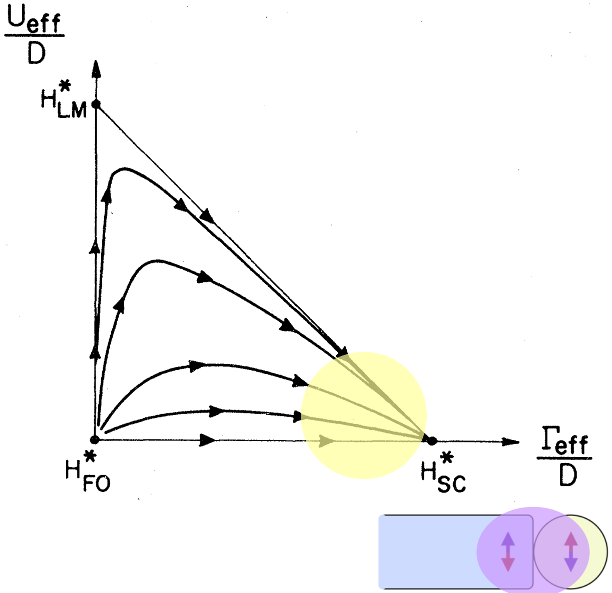
free-orbital regime:
$|\varepsilon^{\mathrm{f}}_{\mathrm{e}\sigma}| < \nu$:



$$H^*_{\mathrm{FO}} = H^{\mathrm{f}}_{\mathrm{QD}} , \quad H'_{\mathrm{FO}} = \sqrt{\frac{\Gamma}{\pi\rho}} \sum_\sigma (e^\dagger_\sigma c_\sigma + \mathrm{h.c.})$$

$$A^{\mathrm{FO}}_\sigma(\nu) = \frac{4\Gamma}{\nu^2} \theta(\nu - |\varepsilon^{\mathrm{f}}_{\mathrm{e}\sigma}|)$$

Strong-Coupling Regime ($T \ll \nu \ll T_K$)



$$H_{\rm SC} = \sum_{k\sigma} \tilde{\varepsilon}_{k\sigma} \tilde{c}^\dagger_{k\sigma} \tilde{c}_{k\sigma}$$

Use analogy to x-ray edge problem: (Mahan '67)

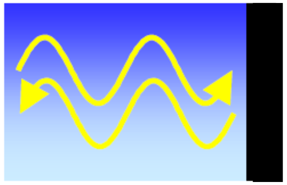
$$A_\sigma(\nu) = -2{\rm Im}\mathcal{G}^\sigma_{\rm ee}(\nu) \sim \nu^{-\eta_\sigma}$$

$$\mathcal{G}^\sigma_{\rm ee}(t) \sim \langle \psi_{\rm i}(0^+) | \psi_{\rm i}(t) \rangle \sim t^{-\eta'_\sigma},$$

$$|\langle \psi_{\rm i}(0^+) | \psi_{\rm i}(\infty) \rangle|^2 \sim N^{-\eta'_\sigma}$$

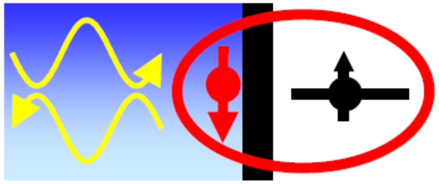
Anderson orthogonality

$|{\rm G}\rangle_{\rm i}$



unperturbed Fermi sea

$|\psi_{\rm i}(\infty)\rangle$



screened
Kondo singlet

Strong-Coupling Regime ($T \ll \nu \ll T_K$)

$$H_{\mathrm{SC}} = \sum_{k\sigma} \tilde{\varepsilon}_{k\sigma} \tilde{c}^{\dagger}_{k\sigma} \tilde{c}_{k\sigma}$$

Use analogy to x-ray edge problem:

$$A_\sigma(\nu) = -2\mathrm{Im}\mathcal{G}^{\sigma}_{\mathrm{ee}}(\nu) \quad \sim \quad \nu^{-\eta_\sigma}$$

$$\mathcal{G}^{\sigma}_{\mathrm{ee}}(t) \sim \langle \psi_{\mathrm{i}}(0^+) | \psi_{\mathrm{i}}(t) \rangle \sim t^{-\eta'_\sigma},$$

$$|\langle \psi_{\mathrm{i}}(0^+) | \psi_{\mathrm{i}}(\infty) \rangle|^2 \sim N^{-\eta'_\sigma}$$

Anderson orthogonality

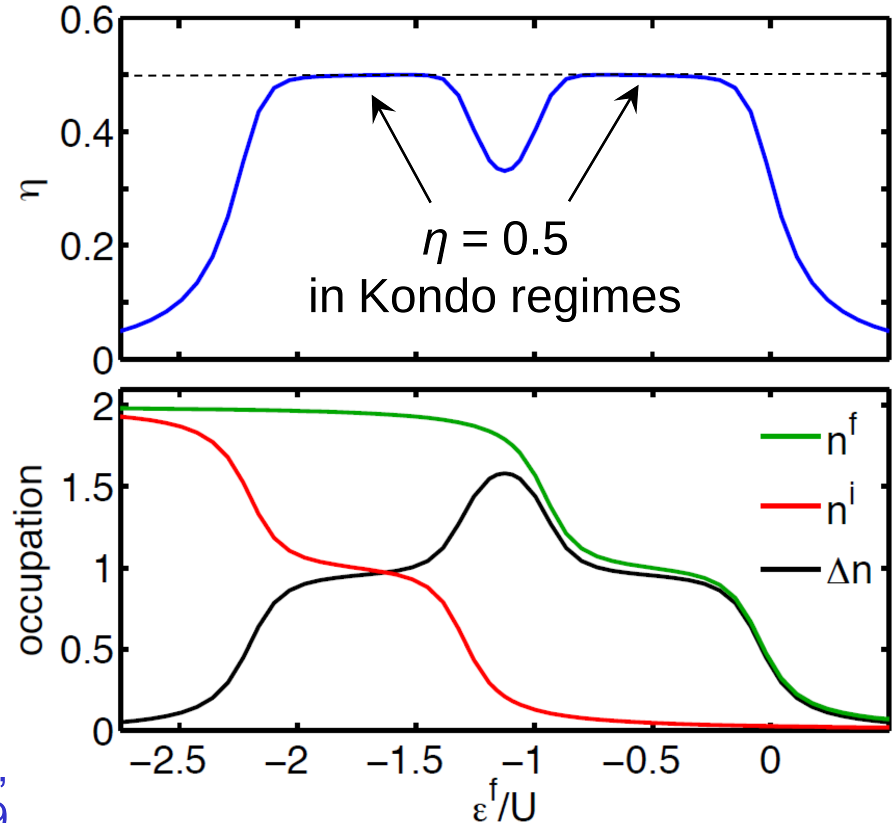
$$\eta_\sigma = 1 - \sum_{\sigma'} (\Delta n'_{\mathrm{e}\sigma'})^2$$

(Friedel, '56, Nozieres, '69, Hopfield '69)

$$\begin{aligned} \Delta n'_{\mathrm{e}\sigma'} &= \langle n_{\mathrm{e}\sigma'} \rangle_\infty - \langle n_{\mathrm{e}\sigma'} \rangle_{0+} \\ &= \underbrace{\langle n_{\mathrm{e}\sigma'} \rangle_{\mathrm{f}} - \langle n_{\mathrm{e}\sigma'} \rangle_{\mathrm{i}}}_{\text{change in local charge}} - \delta_{\sigma'\sigma} \end{aligned}$$

spin symmtery is broken by polarization of incident photon

AO exponent tunable by gate voltage



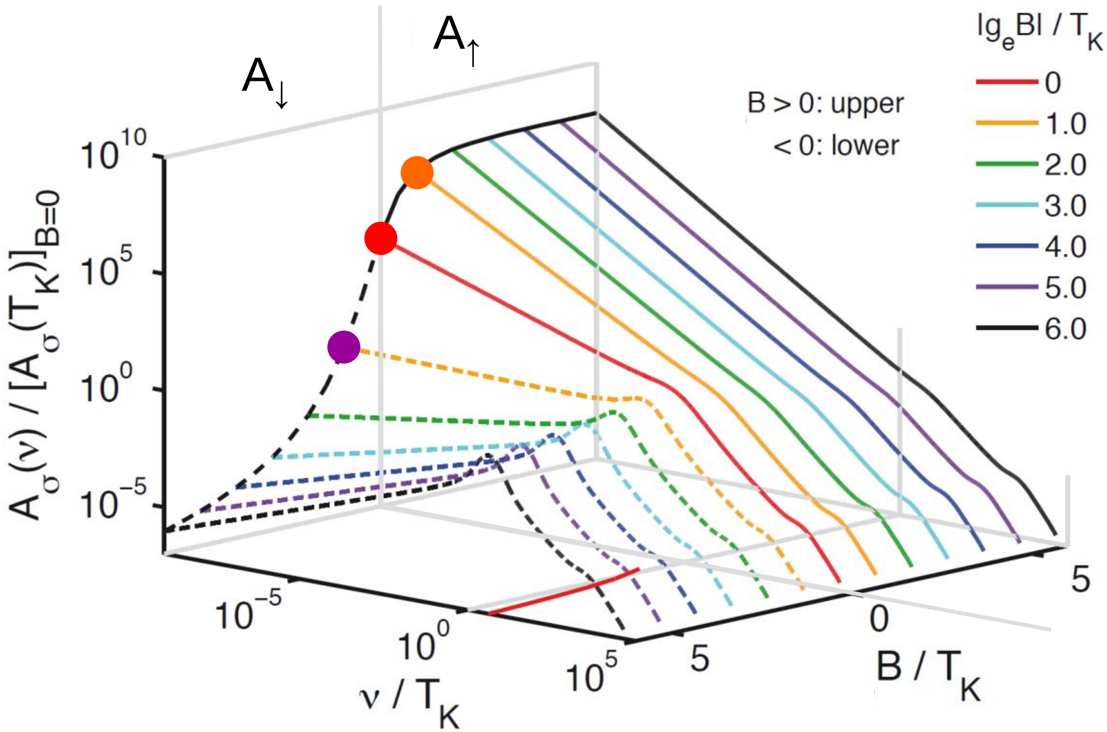
Absorption line shape: B-dependence (SAM)

$$A_\sigma(\nu) \sim \nu^{-\eta_\sigma}$$

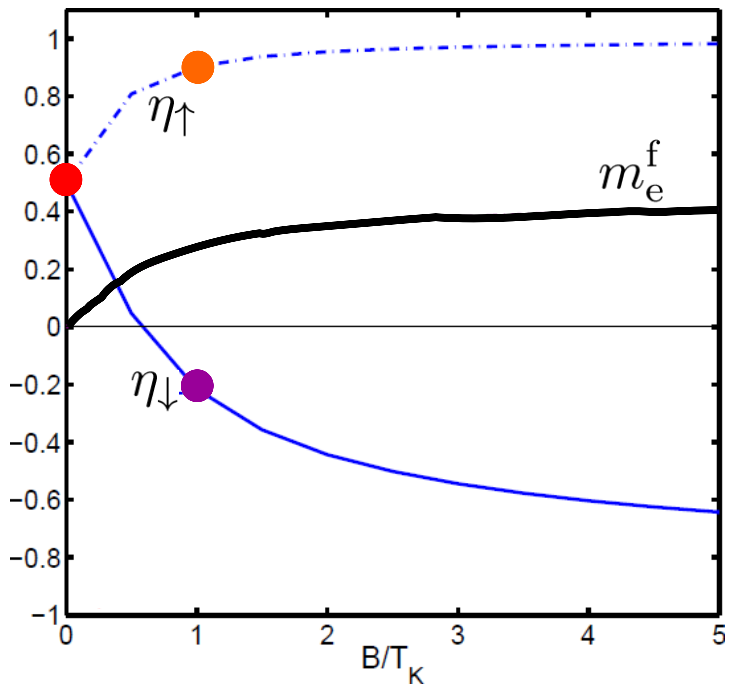
$$\eta_\sigma = \frac{1}{2} + 2\sigma m_e^f - 2(m_e^f)^2$$

final magnetization

Strong asymmetry under reversal of incident polarization for fixed magnetic field



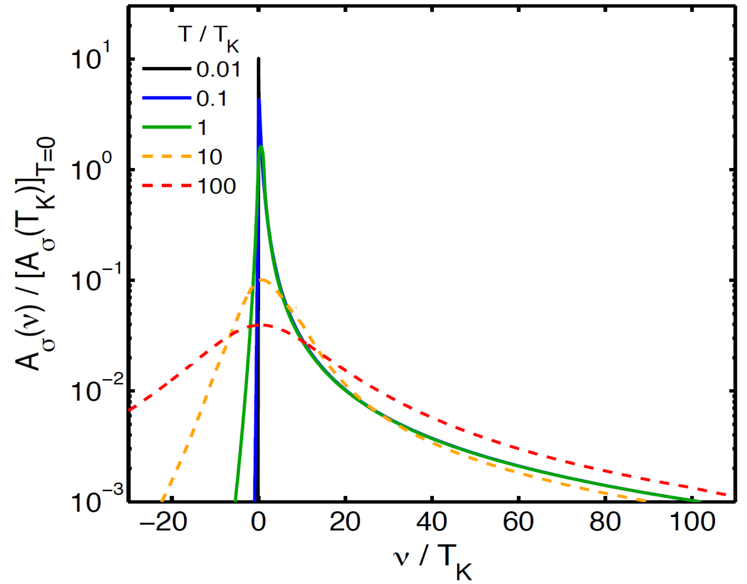
AO exponent tunable by magnetic field



- $A_\uparrow$: less orthogonality, larger matrix element more absorption
- $A_\downarrow$: more orthogonality smaller matrix element less absorption

Main predictions

Absorption spectrum maps out physics of different fixed points



In local moment regime (T < ν < Tk):

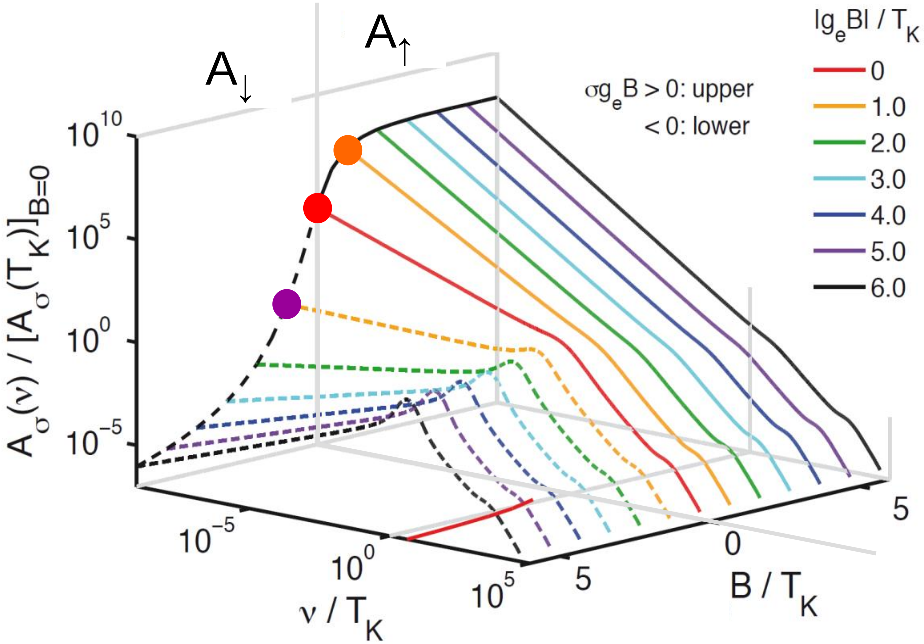
- $$A_\sigma(\nu) \sim \frac{1}{\nu \ln^2(\nu/T_K)}$$

- ν/Tk scaling

In strong-coupling regime (T < ν < Tk):

For T/Tk → 0: powerlaw divergence

$$A_\sigma(\nu) \sim \nu^{-\eta_\sigma}$$



Anderson/Mahan-exponents

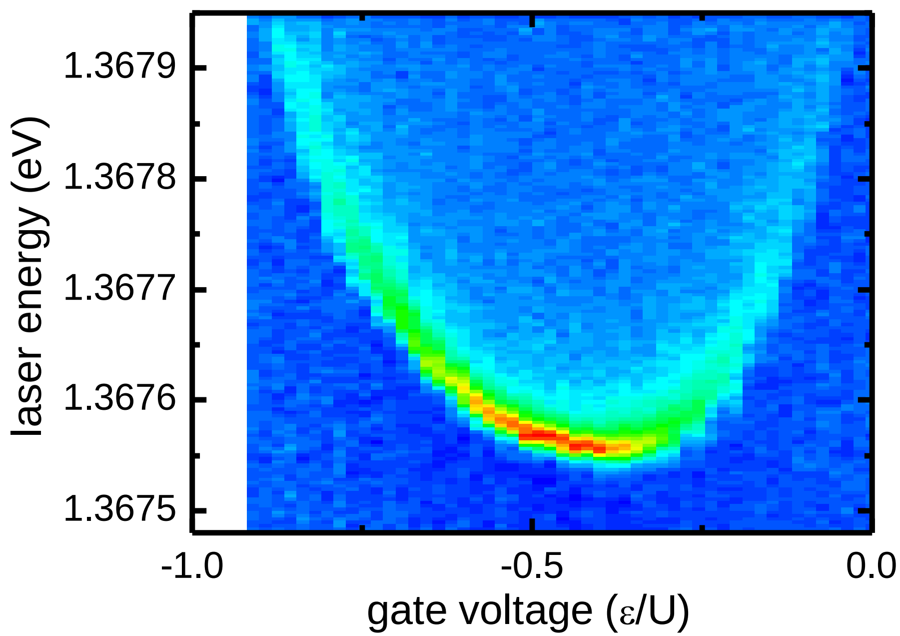
- have universal value η=0.5
  for symmetric Anderson model at B=0;

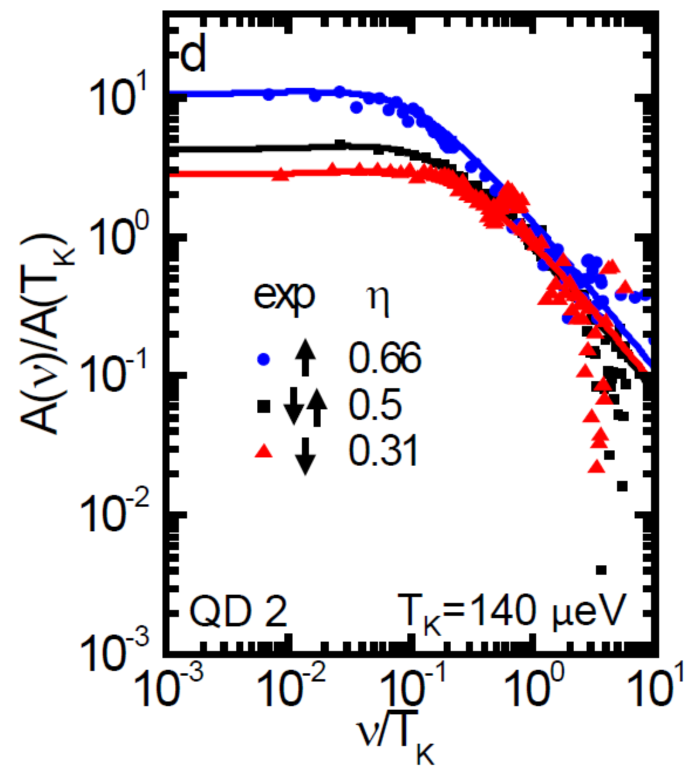
- are tunable by Vg and B

Experiment: Quantum quench of Kondo correlations in optical absorption

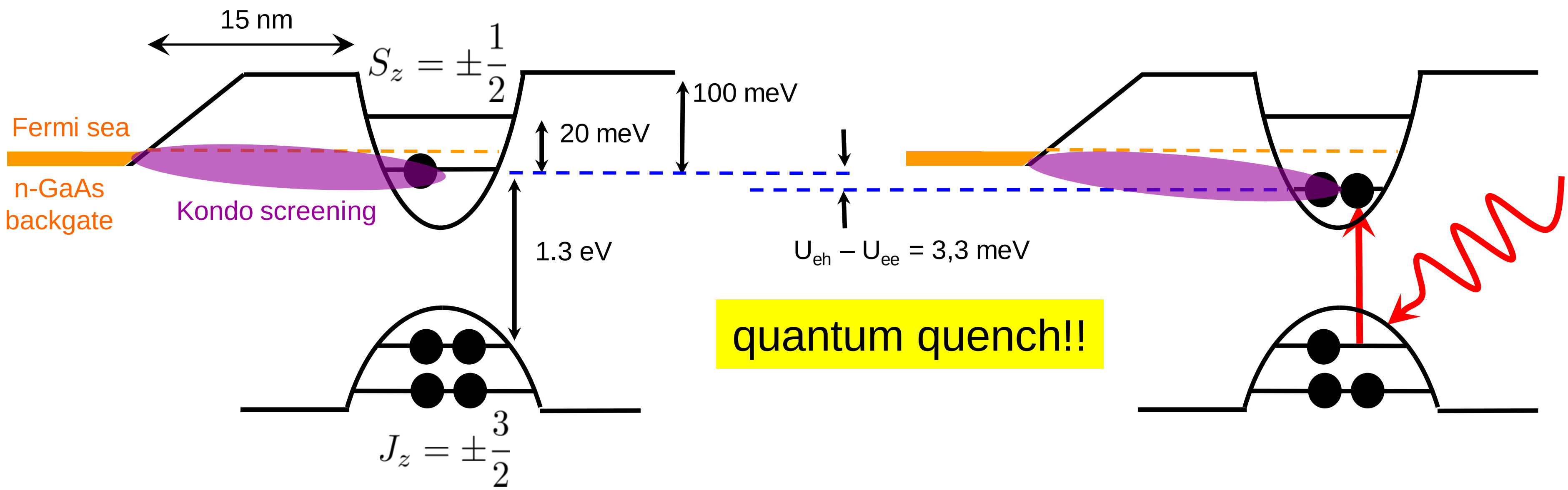
Christian Latta, F. Haupt, P. Fallahi, S. Faelt, A. Imamoglu (ETH), H. Tureci (Princeton)

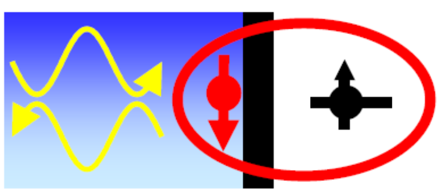
Markus Hanl, Andreas Weichselbaum, Jan von Delft (LMU), Leonid Glazman (Yale)



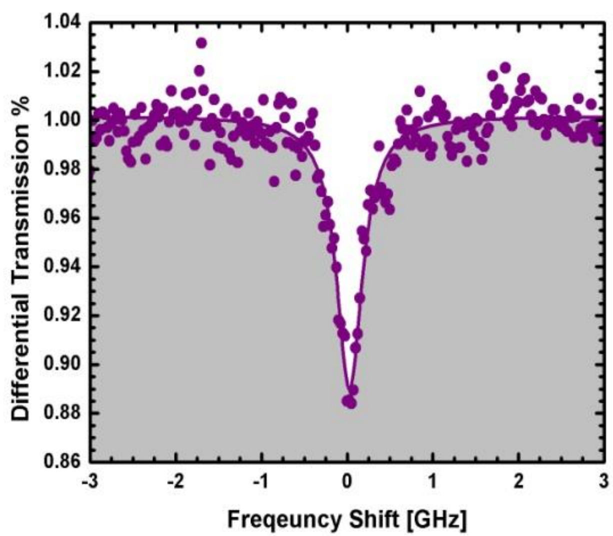


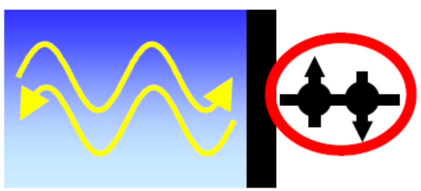
Optical absorption: X^- transition





screened
Kondo singlet

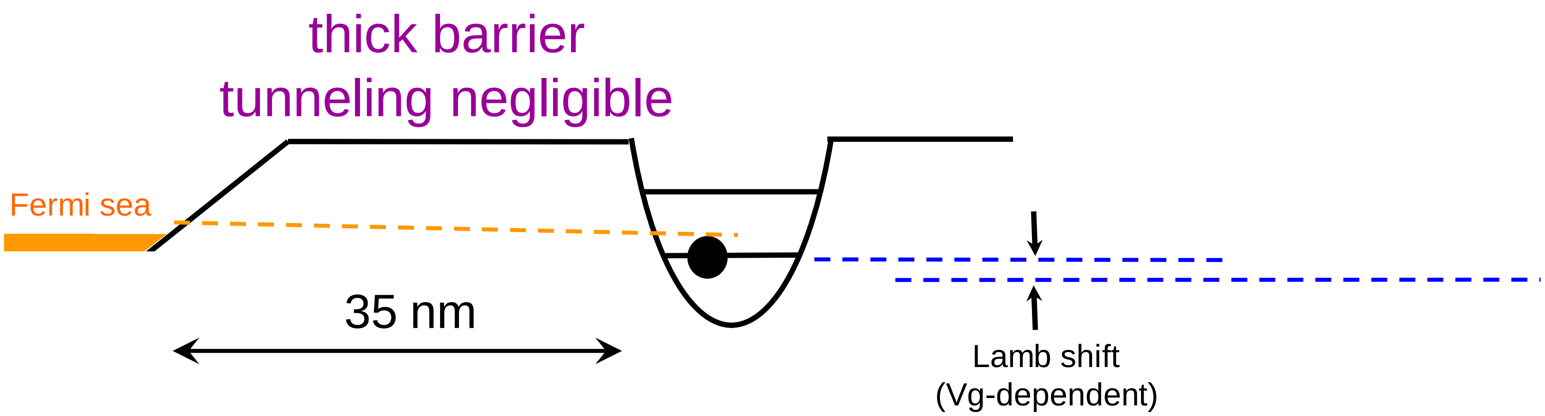


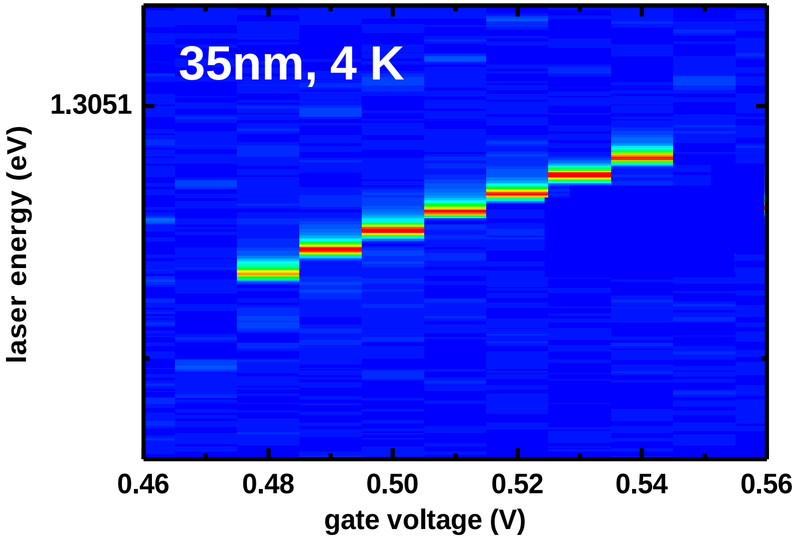


local singlet

tune laser frequency across resonance,
monitor transmitted field intensity

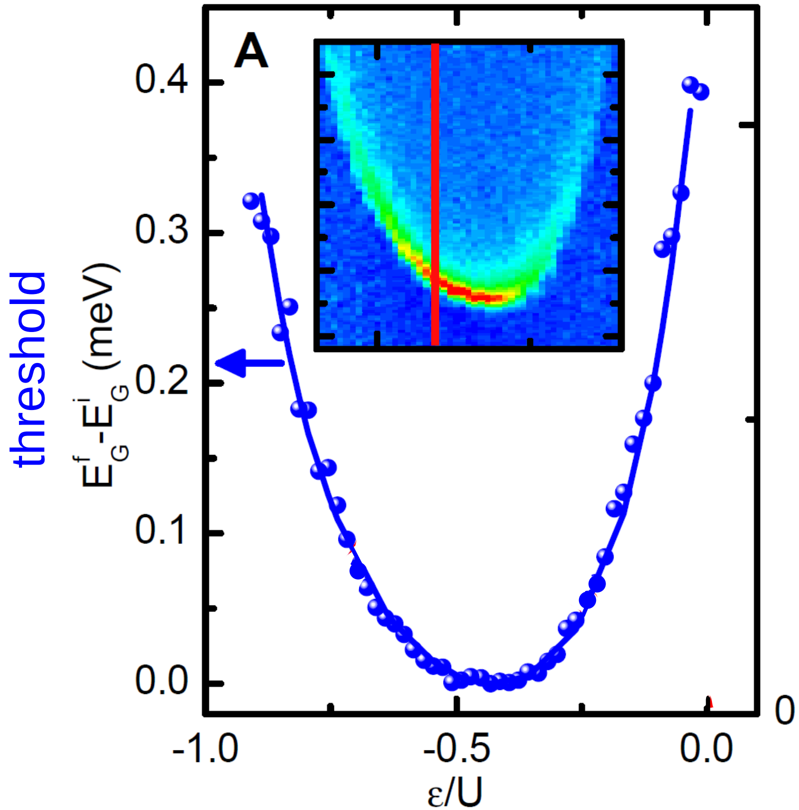
Influence on tunnel barrier width on X- absorption

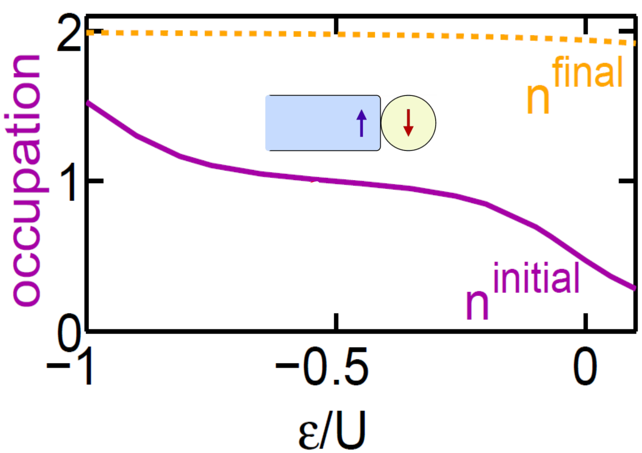




linear dc-Stark shift

Fixing model parameters by fitting NRG to data





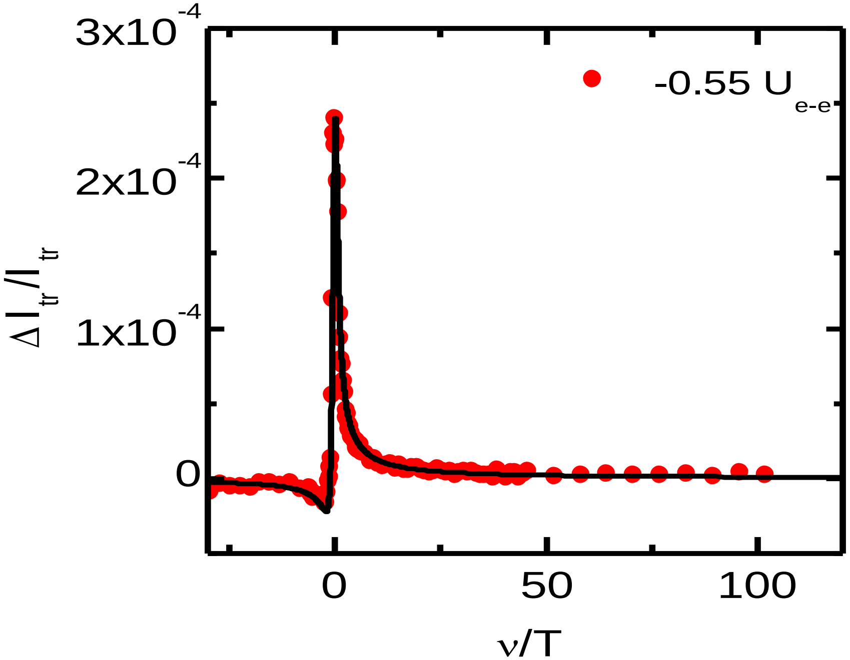
From fit to NRG for threshold:

$U_{e\text{-}h}$ = 11 meV

$U_{e-e} = 7.5\text{meV}$

$\Gamma = 0.7\text{meV}$

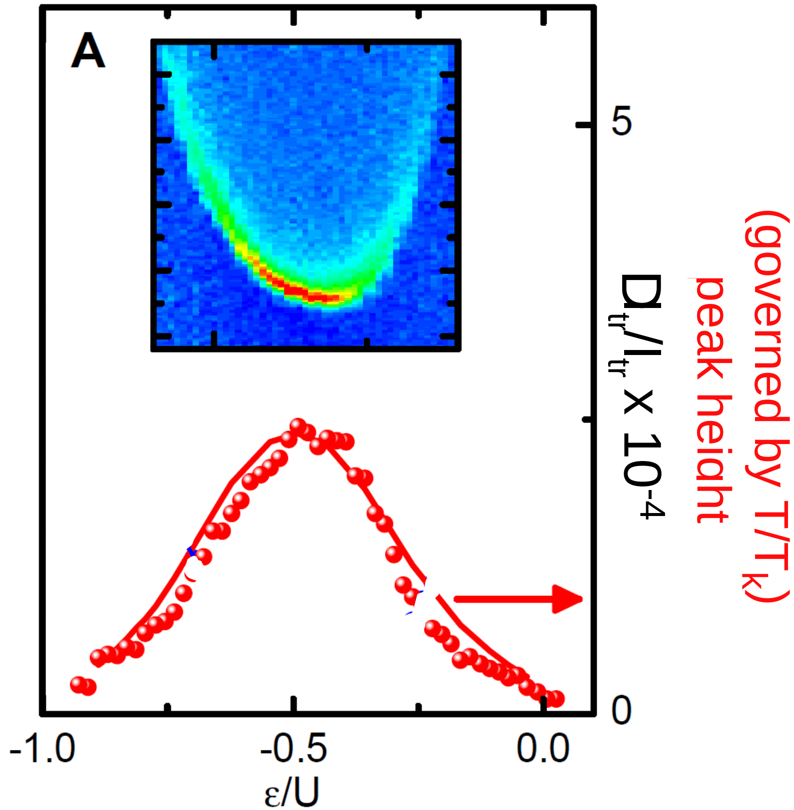
$D = 3.5\text{meV}$

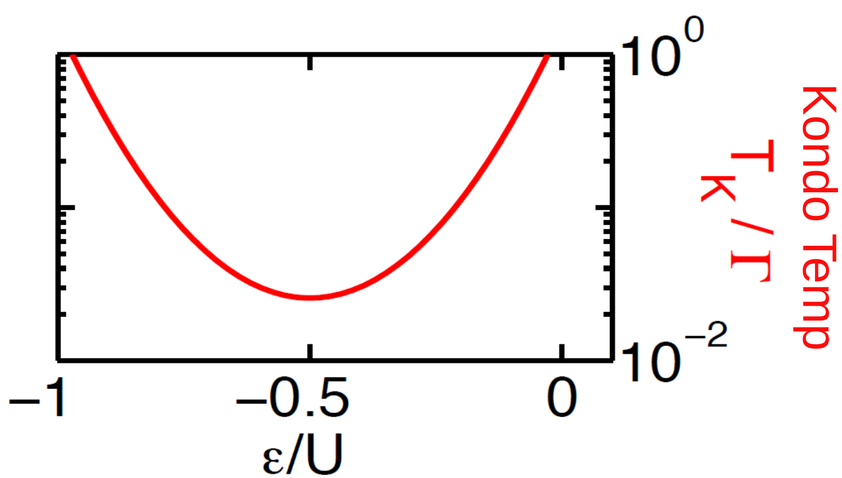


From fit to NRG for $\nu/T < 0$:

T = 180 mK

Fixing model parameters by fitting NRG to data





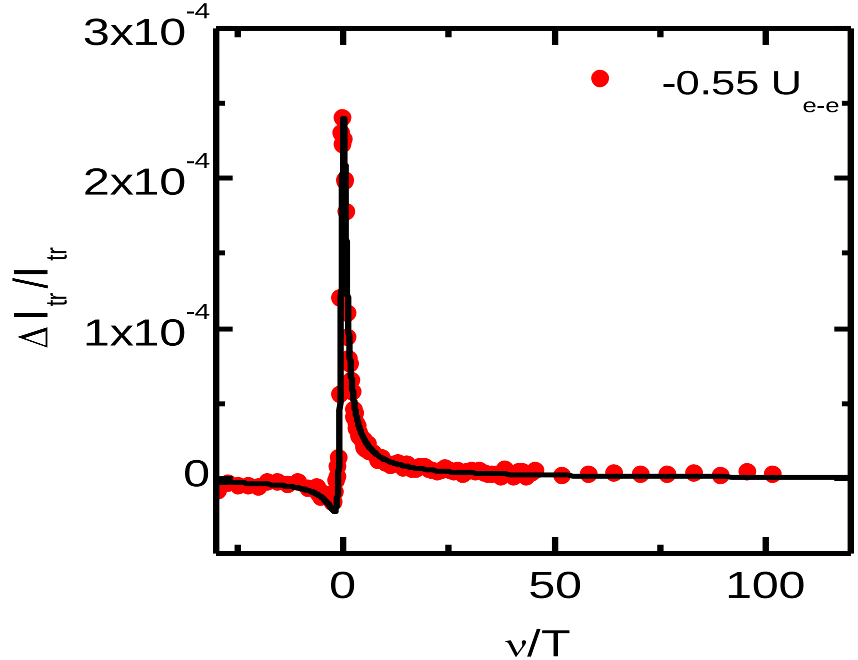
From fit to NRG for threshold:

$U_{e\text{-}h}$ = 11 meV

$U_{e-e} = 7.5\text{meV}$

$\Gamma = 0.7\text{meV}$

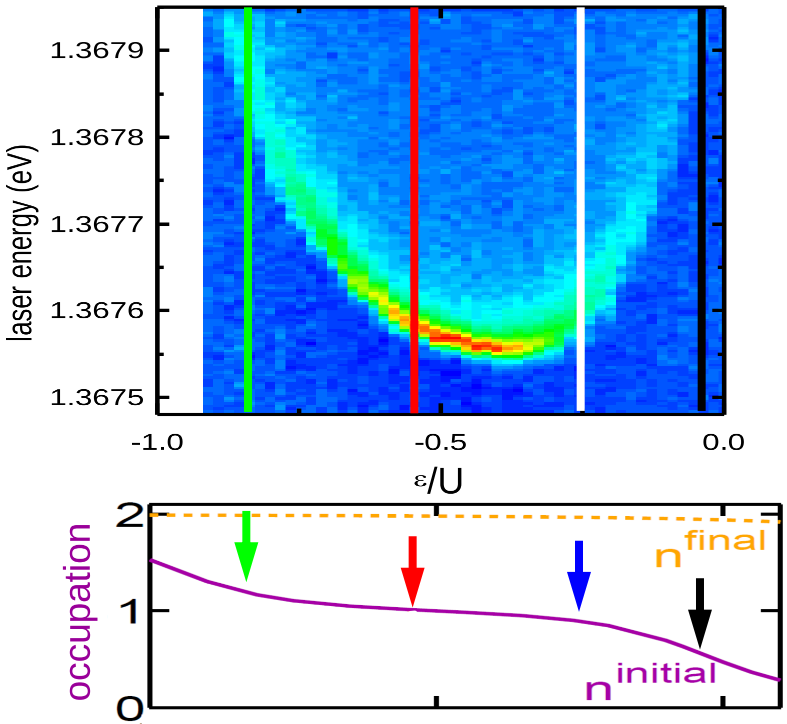
$D = 3.5\text{meV}$



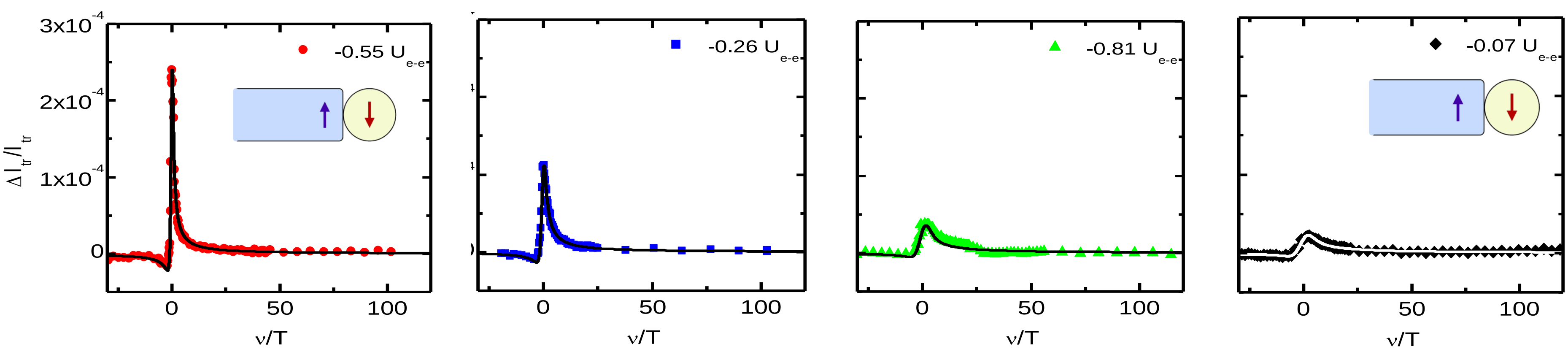
From fit to NRG for $\nu/T < 0$:

T = 180 mK

Anatomy of the line shapes

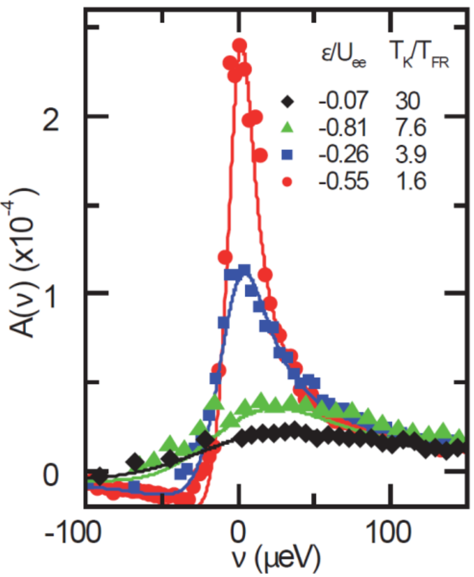


NRG line shapes are no fits!



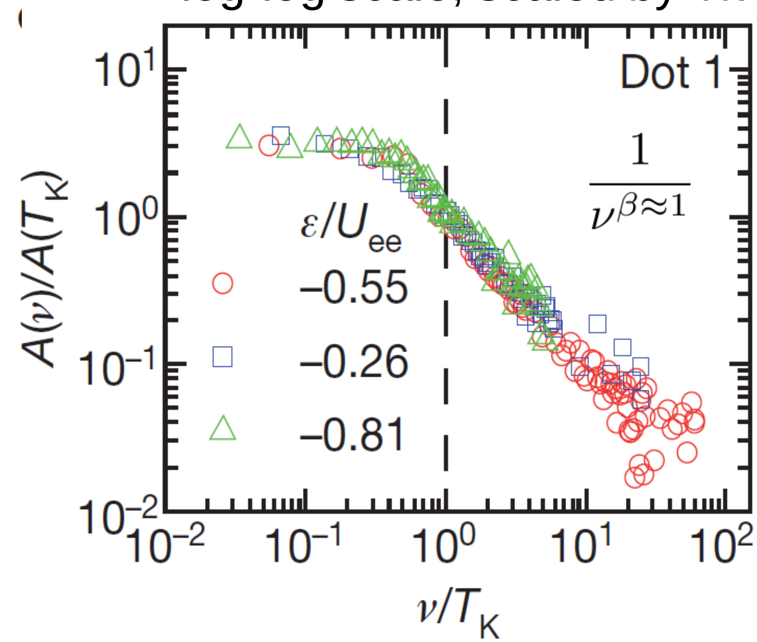
Scaling collapse

linear scale

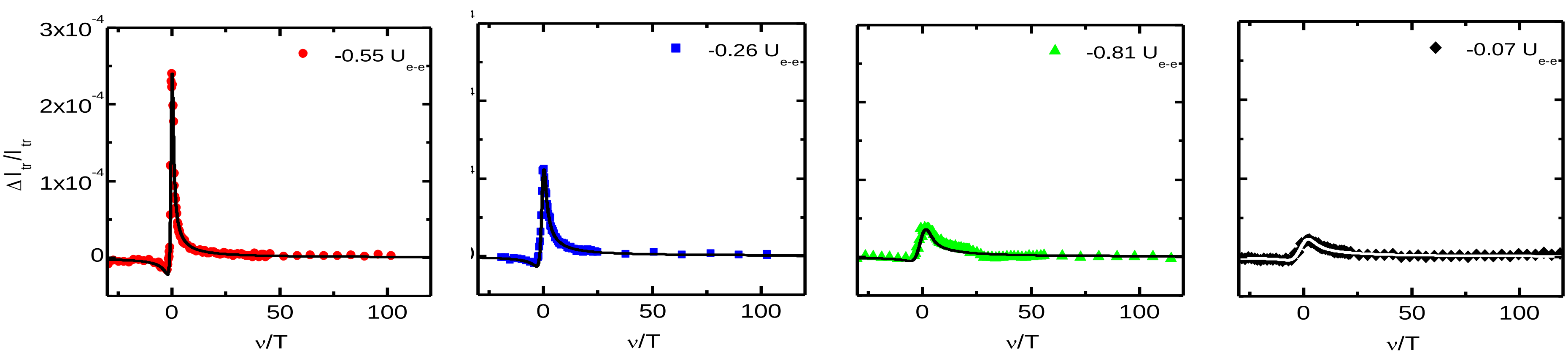


log-log scale

log-log scale, scaled by Tk



scaling collapse: signature of local moment regime!



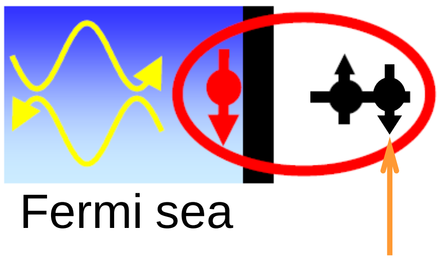
Non-perturbative regime: $T < V < T_K$

Anderson orthogonality catastrophe (AOC)

$|G\rangle_i$

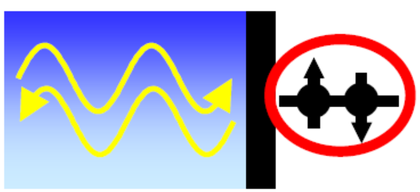
Prediction: AO exponent
tunable by magnetic field

screened Kondo singlet



Fermi sea

$|\psi_i(\infty)\rangle$



local singlet

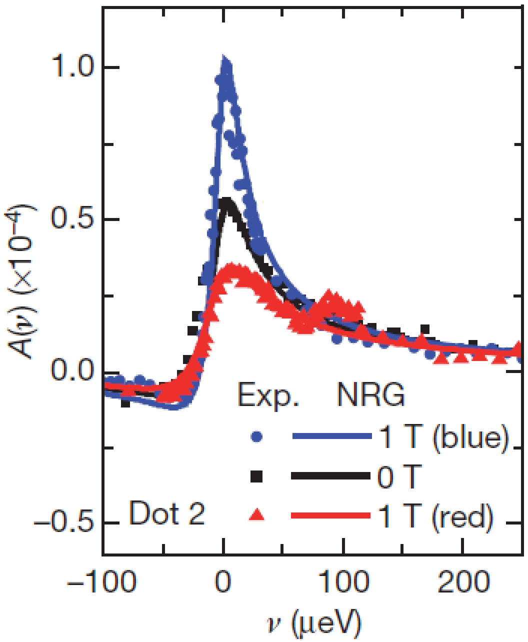
Initial state just after absorption and final state in long-time limit are orthogonal:

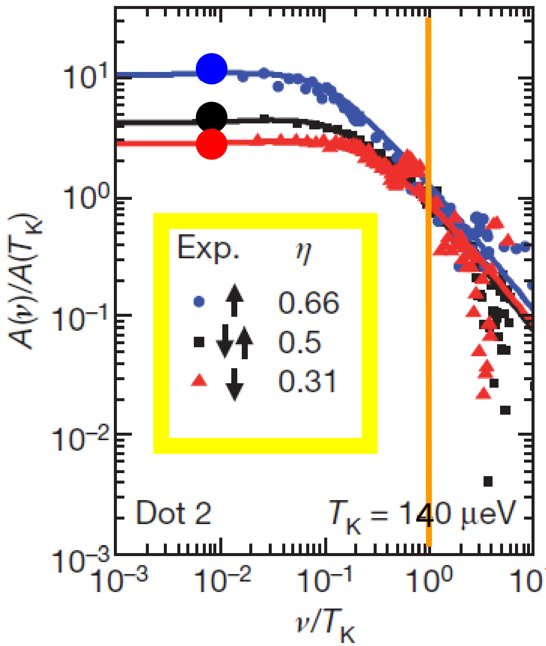
$$|\langle\psi_i(0^+)|\psi_i(\infty)\rangle|^2 \sim N^{-\eta'_\sigma} \qquad A_\sigma(\nu) \sim \nu^{-\eta_\sigma}$$

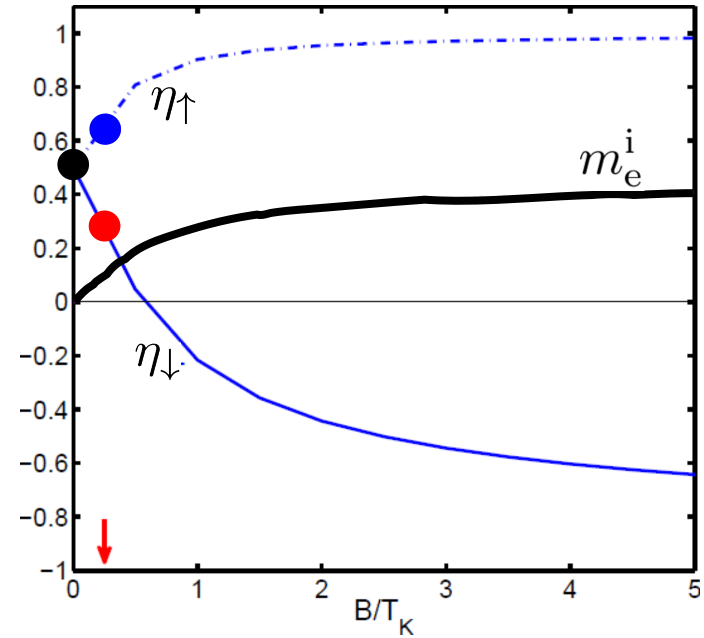
Observation of B-tunable exponents

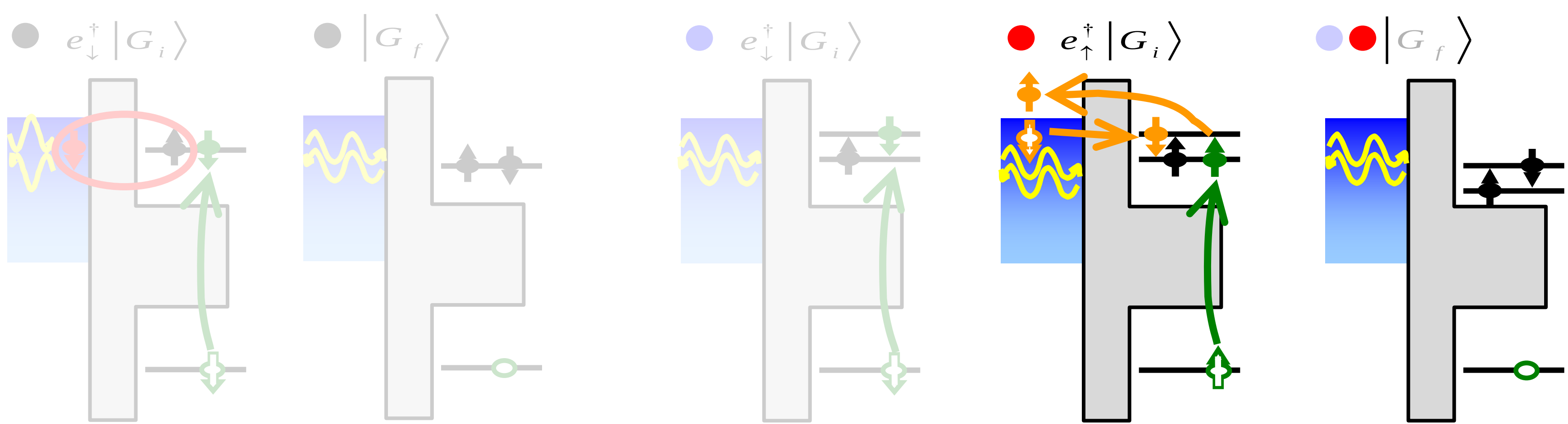
strong-coupling behavior
has been observed!

Strongly coupled QD2 (Tk/T = 9)

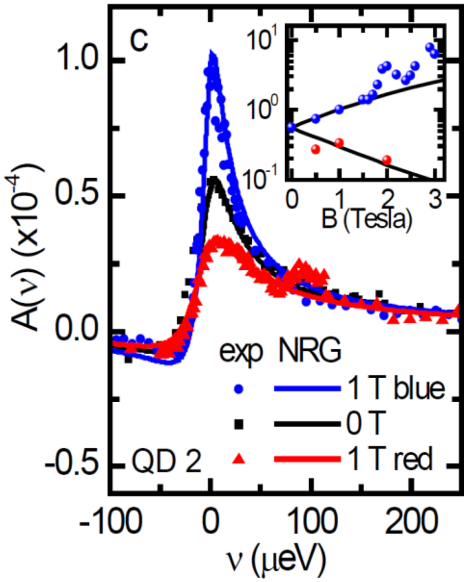








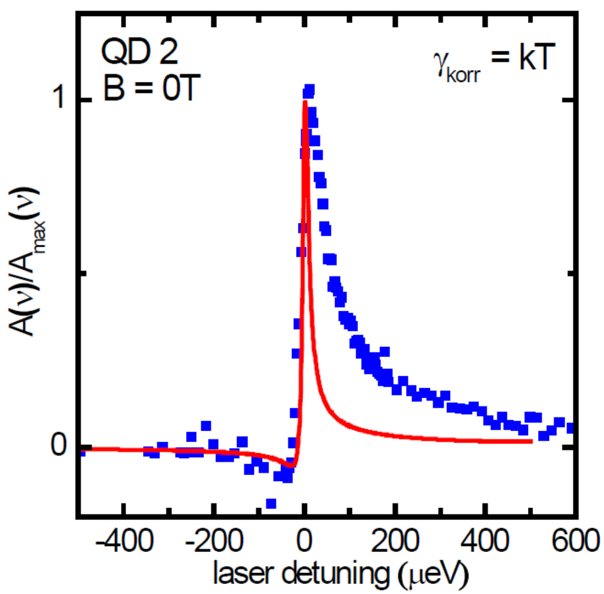
Oscillations of Tk at large fields



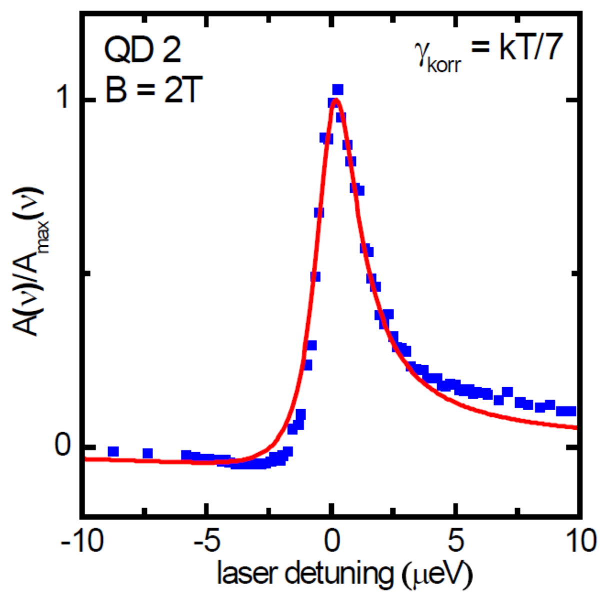
Landau levels produce oscillations in DOS of leads, and hence oscillations in Tk

Perturbative lineshape: $$A(\nu) \propto \frac{\nu/T}{1-e^{-\nu/T}} \frac{\gamma}{\nu^2+\gamma^2/4}$$

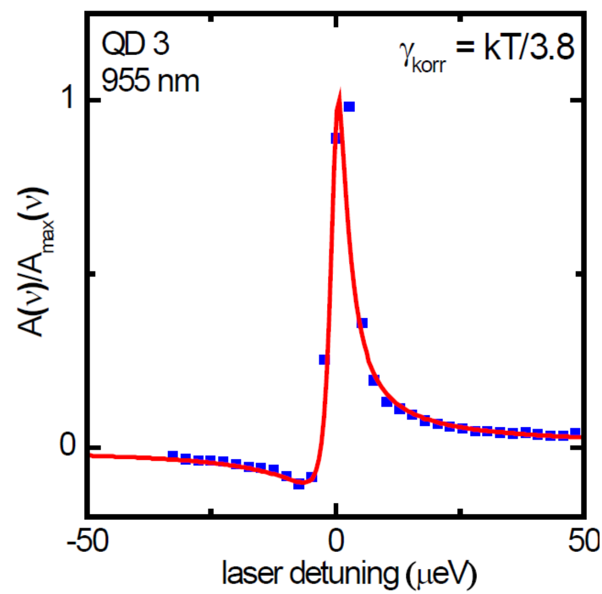
QD2 with large Tk



QD2 with small Tk



QD3: weakly coupled



Main experimental results

Optical signatures of Kondo effect have been observed:

Local moment regime:

- Kondo screening reduces magnetization
- Scaling collapse

Strong-coupling regime:

- Finite temperature hides $\nu^{-\eta}$ behavior,
- but B-tuning of exponents has been observed

NRG reproduces data very well !

Outlook: high-intensity laser = strong driving!