Shedding light on non-equilibrium dynamics of a spin coupled to fermionic reservoir

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(Dated: July 22, 2009)

A single confined spin interacting with a solid-state environment has emerged as one of the fundamental paradigms of mesoscopic physics. In contrast to standard quantum optical systems, decoherence that stems from these interactions can in general not be treated using the Born-Markov approximation at low temperatures. Here we study the non-equilibrium dynamics of a single-spin in a semiconductor quantum dot adjacent to a fermionic reservoir and show how the dynamics can be revealed in detail in an optical absorption experiment. We show that the highly asymmetrical optical absorption lineshape of the resulting Kondo exciton consists of three distinct frequency domains, corresponding to short, intermediate and long times after the initial excitation, which are in turn described by the three fixed points of the single-impurity Anderson Hamiltonian. The zero-temperature power-law singularity dominating the lineshape is linked to dynamically generated Kondo correlations in the photo-excited state. We show that this power-law singularity is tunable with gate voltage and magnetic field, and universal.

Even though quantum dots (QD) are commonly referred to as artificial atoms, their physical properties can be substantially different from that of real atoms. In particular, a single electron spin confined in a QD is subject to hyperfine interactions with the QD nuclear spin ensemble and exchange interactions with the Fermi gas that controls the charging state of the QD. For many applications, such as those aimed at using QD spins to represent quantum information, the nuclear spin ensemble and the Fermi gas of electrons are treated as reservoirs that induce decoherence of the confined electron spin. Tremendous progress in understanding and controlling spin decoherence induced in particular by QD hyperfine interactions has been achieved[1]. In contrast, relatively few studies have addressed the real-time dynamics of an optically excited QD electron coupled to a nearby fermionic reservoir (FR).

The role of coherent tunnel coupling between a QD and a FR has been investigated since the late 1990s in the context of low-temperature transport spectroscopy[2, 3, 4, 5] and is known to lead to one of the most spectacular phenomena of many-body physics – the Kondo effect[6]. The conductance through a QD in the Coulomb blockaded regime is proportional to the local density of states (LDOS) of the QD level; the Kondo effect introduces a quasiparticle peak in the LDOS, the Kondo resonance, at the Fermi energy $\varepsilon_F$, which leads to enhanced linear conductance through the QD. In contrast, optical signatures of the Kondo effect in quantum dots have so far not been observed experimentally[7, 8]. Prior theoretical work on this topic is also scarce, yet point to novel signatures: Govorov and co-workers predicted[9], using a simplified approach based on variational wave functions[10], that doubly charged QDs could exhibit optical resonances whose width depends on an energy scale that is determined by the exchange interactions.

Here we analyze the non-equilibrium dynamics following a quantum quench of a QD spin that is coupled to a FR. In a quantum quench, parameters of the Hamiltonian of an interacting quantum system are changed over a very short time scale. We show that an optical absorption experiment naturally implements such a quantum quench, and the measured optical absorption lineshape reveals, in a uniquely direct way, how for intermediate and long timescales standard quantum optical techniques such as Markov and Born approximations fail to describe the dynamics. Since the absorption of a photon suddenly switches on an attractive potential that (under suitable conditions) favors Kondo correlations and spin screening, the dynamic emergence of such correlations with time can be probed in ways not possible in transport experiments. For short times (large detunings), charge fluctuations dominate and coupling to the FR can be described using the Born-Markov approximation[12]. For intermediate times (intermediate detunings) spin fluctuations dominate; since the spin excitations of the FR are long-lived, the dynamics in this regime is non-Markovian. For long

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times (small detunings), the orthogonality catastrophe physics [13] dominates and leads to a complete breakdown of the Born-Markov approximation. Remarkably, the resulting highly asymmetrical and singular absorption lineshape directly maps out these three regimes, which we link to the three fixed points of the single-impurity Anderson Hamiltonian. We find that the power-law singularity in the lineshape is tunable with gate voltage and magnetic field, and universal.

Model for a tunable, optically active quantum impurity — the excitonic Anderson Model. We consider a QD, tunnel-coupled to a FR (see Fig. 1), whose charge state is controllable via an external gate voltage $V_g$ applied between a top Schottky gate and the FR (see Appendix 1 for details). Assume $V_g$ to be tuned such that optical absorption occurs via the so-called neutral exciton transition [14] ($X^0$), in which an electron-hole pair is created in the localized s-orbitals of the QD’s conduction- and valence bands (to be called e- and h-levels, respectively). For circularly polarized light with frequency $\omega$, propagating along the z-axis of the heterostructure [15], the QD-light interaction is described by $H_L \propto (e^\dagger_x h^\dagger_y e^{i \omega t} + \text{h.c.})$, where $e^\dagger_x / h^\dagger_y$ create an electron/hole in the e- and h-levels, respectively and $s = -\bar{s} \in \{+, -\}$. We model the system before/after absorption by the initial/final Hamiltonian $H^{i/f} = H^{i/f}_{\text{QD}} + H_c + H_t$, where

$$H^{i/f}_{\text{QD}} = \sum_{\sigma} \epsilon_{\text{e} \sigma} n_{\text{e} \sigma} + U n_{\text{e} \uparrow} n_{\text{e} \downarrow} + \delta_{\text{ad}} \epsilon_{\text{h} \bar{\sigma}}$$

(a = i, f) (1)

describes the QD, with Coulomb cost $U$ for double occupancy of the e-level, $n_{\text{e} \sigma} = e^\dagger_x e_{\sigma}$, and hole energy $\epsilon_{\text{h} \bar{\sigma}}$ ($> 0$, on the order of the band gap). The e-level’s initial and final energies before and after absorption, $\epsilon_{\text{e} \sigma}^a$ (a = i, f), differ by the Coulomb attraction $U_{\text{eh}} (> 0)$ between the newly created electron-hole pair, which pulls the final e-level downward, $\epsilon_{\text{e} \sigma}^f = \epsilon_{\text{e} \sigma} - \delta_{\text{ad}} U_{\text{eh}}$ (Fig. 1b). This stabilizes the excited electron against decay into the FR, provided that $\epsilon_{\text{e} \sigma}^f$ lies below the FR’s Fermi energy $\epsilon_F = 0$. $H_0 = \sum_{k \sigma} \epsilon_{k \sigma} c^\dagger_{k \sigma} c_{k \sigma}$ represents a noninteracting conduction band (the FR) with half-width $D = 1/(2p)$ and constant density of states $\rho(\epsilon_k) = p(D - |\epsilon_k|)$ per spin, while $H_t = \sqrt{1/\rho} \sum \epsilon_{k \sigma}(e^\dagger_{k \sigma} c_{\sigma} + \text{h.c.})$, with $c_{\sigma} = \sum_k c_{k \sigma}$, describes its tunnel-coupling to the e-level, giving it a width $\Gamma$. A magnetic field $B$ along the growth-direction of the heterostructure (Faraday configuration) causes a Zeeman splitting, $\epsilon_{\text{e} \sigma} = \epsilon_e + \sigma g_B B = \epsilon_e + \sigma g_B B$ (see Appendix 1; the Zeeman splitting of FR states can be neglected for our purposes, see Appendix 8.). We set $\mu_B = h = k_B = 1$, give energies in units of $D = 1$ throughout, and assume $T, B \ll \Gamma \ll U, U_{\text{eh}} < D \ll \epsilon_{\text{e} \bar{\sigma}}$. A realistic set of parameters would be, e.g., $\Gamma \approx 1-10$ meV, $U \approx U_{\text{eh}} \approx 15-25$ meV (in general $U_{\text{eh}}$ is slightly larger than $U$, resulting in a “tronic redshift” of the order of $\approx 5$ meV), $D \approx 30$ meV, $\epsilon_{\text{e} \bar{\sigma}} \approx 1.3$ eV, $g_e \approx -0.6-0.7$, $g_h \approx 1.1-1.2$.

We focus on the case where the e-level is essentially empty in the initial state and singly-occupied in the ground state of the final Hamiltonian, $n_{\text{e} \bar{\sigma}}^0 \approx 0$ and $n_{\text{e} \bar{\sigma}}^f \approx 1$. (Here $\tilde{n}_{\text{e} \sigma} = \langle n_{\text{e} \sigma} \rangle$ is the thermal average with respect to $H^f$ of $n_{\text{e} \sigma} = \sum_{\sigma} n_{\text{e} \sigma}$.) This requires $\epsilon_{\text{e} \sigma}^f \gg \Gamma$, and $|\epsilon_{\text{e} \sigma}^f| < |\epsilon_{\text{e} \sigma}^i| < \Gamma$. The initial ground state, needed below, will thus be approximated by the free Fermi sea, $|G\rangle_i \approx \prod_{\bar{\sigma}} \int e_{\text{e} \bar{\sigma}}^\dagger |\text{Vac}\rangle$, neglecting terms of order $\Gamma/\epsilon_{\text{e} \bar{\sigma}}$. In particular, some (but not all) parts of the text will focus on the case that $H^f$ represents the symmetric excitonic Anderson model ($H^f=\text{SEAM}$), with $\epsilon_{\text{e} \bar{\sigma}}^f = -\epsilon_{\text{e} \bar{\sigma}}^i = -U/2$, for which $n_{\text{e} \bar{\sigma}}^i = 1$ exactly.

Time evolution of the charge and spin after a quantum quench induced by absorption. To gain intuition for how the system would respond to the sudden creation of an $e^\dagger_x h^\dagger_y$ exciton at time $t = 0$, it is instructive to calculate the subsequent time evolution of the average charge $\bar{n}_{\text{e} \bar{\sigma}}(t) = \langle n_{\text{e} \bar{\sigma}} \rangle + \langle n_{\text{e} \bar{\sigma}} \rangle$ and spin $\bar{\sigma}_{\text{e} \bar{\sigma}}(t) = \frac{1}{2}(\bar{n}_{\text{e} \uparrow} - \bar{n}_{\text{e} \downarrow})$ of the e-level, where $\bar{n}_{\text{e} \bar{\sigma}}(t) = \text{Tr}(e^{-iH^f t} \rho^i e^{iH^f t} |\bar{n}_{\text{e} \bar{\sigma}}\rangle \langle \bar{n}_{\text{e} \bar{\sigma}}|)$ and $\rho^i_{\text{e} \bar{\sigma}} = e_{\text{e} \bar{\sigma}}^i \rho_{\text{e} \bar{\sigma}} / (1 - \bar{n}_{\text{e} \bar{\sigma}})$ is a projected version of the initial density matrix, normalized such that $\bar{n}_{\text{e} \bar{\sigma}}(0) = 1$. Fig. 2 shows a typical result for $T = 0$ and $H^f=\text{SEAM}$, obtained using time-dependent NRG [12] (see Methods and Appendix 3). The non-equilibrium dynamics following such a quantum quench shows two distinct time scales: (i) Fluctuations in both charge and spin set in around the time scale $t \approx 1/|\epsilon_{\text{e} \bar{\sigma}}^i|$ associated with virtual transitions of electrons between e-level and FR. Whereas the charge equilibrates (towards 1) shortly thereafter, (ii) the spin decays (towards $\approx 0$) much more slowly, on the scale $t \approx 1/T_K$, where $T_K = \sqrt{|U|/2e^{-\pi i (\epsilon_{\text{e} \uparrow} + \epsilon_{\text{e} \downarrow})/(2D)}}$ is the Kondo temperature [17, 18] associated with $H^f$. (For finite temperatures, the time scale on which $\bar{n}_{\text{e} \bar{\sigma}}(t)$ decays is $\min\{1/\gamma_{\text{Kor}}, 1/T_K\}$, where $\gamma_{\text{Kor}} = T/\hbar^2 (T/T_K)$ is the Korringa relaxation rate.) The decay is due to spin-flip processes, mediated by electrons of opposite spin hop-
means that for cloud builds up over time, which ultimately screens the energy gap between two e-level spin-flips. As a result a FR screening between e-level and FR, leading to non-Markovian dynamics can be more clearly discerned by measuring the non-equilibrium time-evolution of charge and spin degrees of freedom of the photo-excited electron after the sudden creation of an $\epsilon^a_0 \hbar^\dagger$, exciton at time $t = 0$. While fluctuations in both e-level’s total charge $\bar{n}_e$ and spin set in around the time scale $1/|\epsilon^e_0|$, the equilibration of the spin-$\sigma$ populations $\bar{n}_{e\sigma}(t)$ and screening of the local spin $\bar{m}_e(t)$ sets in on the time scale $1/T_K$. The deviations (by about 3%) of $\bar{n}_e(\infty)$, $\bar{n}_{e\sigma}(\infty)$ and $\bar{m}_e(\infty)$ from their expected equilibrium values ($1, \frac{1}{2}$ and 0, respectively), for the case $H^f$=SEAM depicted here), are known artifacts of time-dependent NRG (see Appendix 3). Here, $t$ is measured in units of $1/D$. NRG parameters: $U = 0.1 D$, $\epsilon^e_0 = 0.75 U$, $\epsilon^f_0 = -0.5 U$, $\Gamma = 0.03 U$, $T_K = 5.9 \cdot 10^{-9} \Gamma$, $T = 0$, $B = 0$, $\Lambda = 1.8$, Kept states: 1024, $\alpha = 0.4$.

![FIG. 2: Non-equilibrium time evolution of spin and charge of the photexcited electron. Plotted is the non-equilibrium time-evolution of charge and spin degrees of freedom of the photexcited electron after the sudden creation of an $\epsilon^a_0 \hbar^\dagger$, exciton at time $t = 0$. While fluctuations in both e-level’s total charge $\bar{n}_e$ and spin set in around the time scale $1/|\epsilon^e_0|$, the equilibration of the spin-\(\sigma\) populations $\bar{n}_{e\sigma}(t)$ and screening of the local spin $\bar{m}_e(t)$ sets in on the time scale $1/T_K$. The deviations (by about 3%) of $\bar{n}_e(\infty)$, $\bar{n}_{e\sigma}(\infty)$ and $\bar{m}_e(\infty)$ from their expected equilibrium values ($1, \frac{1}{2}$ and 0, respectively), for the case $H^f$=SEAM depicted here), are known artifacts of time-dependent NRG (see Appendix 3). Here, $t$ is measured in units of $1/D$. NRG parameters: $U = 0.1 D$, $\epsilon^e_0 = 0.75 U$, $\epsilon^f_0 = -0.5 U$, $\Gamma = 0.03 U$, $T_K = 5.9 \cdot 10^{-9} \Gamma$, $T = 0$, $B = 0$, $\Lambda = 1.8$, Kept states: 1024, $\alpha = 0.4$.]

As a result a FR screening cloud builds up over time, which ultimately screens the localized spin into a singlet. Note that this in particular means that for $t > 1/T_K$ the “reservoir” (FR) is substantially modified, implying the complete breakdown of the Born-Markov approximation. We will come back to this point further below.

The time-evolution depicted in Fig. 2 could in principle be observed by a $\pi$-pulse excitation of the QD followed by polarization resolved detection of the photoluminescence. However, the fingerprints of the non-equilibrium dynamics can be more clearly discerned by measuring the absorption lineshape of a continuous-wave laser field, as we show next.

Absorption lineshape of a Kondo exciton. Absorption sets in once $\omega_L$ exceeds a threshold frequency $\omega_{1h} = E^f_G - E^f_G$, which is on the order of $\epsilon_{e\sigma} + \epsilon_{h\sigma}$ (minus corrections due to tunneling and correlations). By Fermi’s golden rule the absorption lineshape at temperature $T$ and detuning $\nu = \omega_L - \omega_{1h}$ is proportional to

$$A_\sigma(\nu) = 2\pi \sum_m \rho^m_{ee} \left| \langle n | \epsilon^e_0 | m \rangle \right|^2 \delta(\omega_L - E^f_a + E^f_m) ,$$  \hspace{1cm} (2)

Here $|m\rangle_a$ and $E^f_m$ are the exact eigenstates and -energies of $H^a$ and $\rho^m_{ee} = e^{-E^f_m/T}/Z^*$ the initial Boltzmann weights. For future reference, we note that Eq. (2) can be expressed as $A_\sigma(\nu) = -2\text{Im}G^\sigma_{ee}(\nu)$, where, for $T = 0$,

$$G^\sigma_{ee}(\nu) = i \langle G | \sigma \frac{1}{\nu_+ - H^f_\sigma | G \rangle} ,$$  \hspace{1cm} (3)

with $\nu_+ = \nu + i0$ and $H^f = H^f - E^f_G - \omega_{1h}$. Moreover, the Fourier representation $G^\sigma_{ee}(\nu) = \int dt e^{i\nu t} G^\sigma_{ee}(t)$, where $G^\sigma_{ee}(t) = -i \theta(t) \langle G | \epsilon | H^f \epsilon e^{-iH^f t} | \epsilon \rangle | G \rangle$, makes explicit that this correlator directly probes the dynamics, described above and in Fig. 2, of a photo-generated electron coupled to a FR (see Appendix for details).

We used NRG to calculate $A_\sigma(\nu)$ from Eq. (2), generalizing the approach of Ref. [11] to $T \neq 0$ by following Ref. [19] (see Methods and Appendix 2 for details). For clarity, we focus first on $H^L$=SEAM with $B = 0$. Fig. 3a shows a typical result: As temperature is gradually reduced, an otherwise symmetric lineshape develops into a highly asymmetric one, dramatically increasing in peak-height as $T \to 0$. At $T = 0$, the lineshape displays a threshold behavior, vanishing for $\nu < 0$ and diverging as $\nu$ tends to 0 from above. Fig. 3b analyzes this divergence on a log-log plot, for the case that $T$, which cuts off the divergence, is smaller than all other relevant energy scales (hence $T = 0$ in all analytical calculations below). Three distinct functional forms are discernible in the regimes of “large”, “intermediate” or “small” detuning, labeled (for reasons discussed below) FO, LM and SC, respectively:

\begin{align*}
(\text{FO}) \quad |\epsilon^f_{e\sigma}| \lesssim \nu \lesssim D & : A \propto \nu^{-2} \theta(\nu - |\epsilon^f_{e\sigma}|) ; & (4a) \\
(\text{LM}) \quad T_K \lesssim \nu \lesssim |\epsilon^f_{e\sigma}| & : A \propto \nu^{-1} \ln^{-2}(\nu/T_K) ; & (4b) \\
(\text{SC}) \quad T \lesssim \nu \lesssim T_K & : A \propto \nu^{-\eta_T} . & (4c)
\end{align*}

A central goal of this paper is to explain the remarkable series of cross-overs described above. To this end we note that absorption at large, intermediate or small detuning probes excitations at successively smaller energy scales, corresponding to ever longer time scales after absorption, for which $H^f$ can be represented by expansions $H^f_{\nu=0}$ around the three well-known fixed points of AM: the free orbital, local moment and strong-coupling fixed points ($r = \text{FO}, \text{LM}, \text{SC}$), characterized by charge fluctuations, spin fluctuations and spin screening, respectively.

Large and intermediate detuning dependence of the lineshape – perturbative regime. For large detuning, probing the time interval $t \lesssim 1/|\epsilon^f_{e\sigma}|$ immediately after absorption, the e-level appears as a free, filled orbital perturbed by charge fluctuations, described by the fixed point Hamiltonian $H^f_{\text{FO}} = H_c + H^f_{\text{QD}}$ and the relevant perturbation $H^f_{\text{FO}} = H_t$. Intermediate detuning
probes the times $1/|e_{\sigma e}| \lesssim t \lesssim 1/T_K$ for which real charge fluctuations have frozen out, resulting in a stable local moment (assuming $\bar{n}_e \approx 1$); however, virtual charge fluctuations still cause the local moment to undergo spin fluctuations, which are not yet screened. This is described by $H_{\rm LM} = H_e + \text{const.}$ and the RG-relevant perturbation $H_{\rm LM} = \frac{J}{3} \hat{s}_e \cdot \hat{s}_c$ (a potential scattering term in $H_{\rm LM}$, being RG-irrelevant, will be neglected in the discussion of the intermediate-detuning regime). Here $s_j = \frac{1}{2} \sum_{\sigma\sigma'} \sigma_j^{\sigma\sigma'} \rho_{\sigma\sigma'}$, for $j = e, c$, are spin-operators for the $e$-level and conduction band, respectively ($\hat{r}$ are Pauli matrices), and $J = 2UT/|\pi e (\epsilon^f_e + U)|$ is an effective dimensionless exchange constant.\cite{18} Constant contributions to $H^*_e$ will not be specified, since they affect only $\omega_{\text{th}}$, whose precise value is not of present interest. It suffices to note that for both $r = \text{FO}$ and $\text{LM}$, $e_{\sigma e}^f(G_1)$ is an eigenstate of $H^*_e$ with eigenvalue $\omega_{\text{th}} + E_{\sigma e}$ (within the accuracy of $H^*_e$).

For $r = \text{FO}$ and $\text{LM}$, $A_\sigma(\nu)$ can be calculated using fixed point perturbation theory in $H^*_e$, as outlined in Methods and Appendix 5. At $T = 0$, we find

$$A^{\text{FO}}_{\sigma}(\nu) = \frac{4\Gamma}{\nu^2} \theta(\nu - |\epsilon^f_{\sigma e}|) \quad (|\epsilon^f_{\sigma e}| \lesssim \nu \lesssim D), \quad (5)$$

$$A^{\text{LM}}_{\sigma}(\nu) = \frac{3\pi J^2(\nu)}{4\nu} \quad (T_K \lesssim \nu \lesssim |\epsilon^f_{\sigma e}|). \quad (6)$$

The crossover from LM to FO shows up in our numerical results as a small shoulder or side peak in $A_\sigma(\nu)$ at $|\epsilon^f_{\sigma e}|$, depending on the detailed choice of parameters (easily seen from $A^{\text{LM}}_{\sigma}(\epsilon^f_{\sigma e}) \neq A^{\text{FO}}_{\sigma}(\epsilon^f_{\sigma e})$). This cross-over reflects the fact that for $\nu > |\epsilon^f_{\sigma e}|$, transitions from the h-level into unfilled states of the FR are possible, mediated by $H^*_{\text{FO}}$ and using the e-level (Lorentzian-broadened by charge fluctuations, hence the $\nu^{-2}$ dependence) as intermediate state, without creating additional particle-hole excitations. For intermediate detuning (LM) this is not possible: instead, absorption into the e-level is accompanied by the $H^*_{\text{LM}}$-induced creation of electron-hole pairs in the FR, yielding an additional phase space factor $\propto \nu$. In Eq. (6), we evoked scaling arguments\cite{18} to replace the bare exchange constant $J$ by its renormalized, scale-dependent version, $J(\nu) = \ln\left(\nu/T_K\right).$ Eqs. (5) and (6) reproduce Eqs. (4a) and (4b), and quantitatively agree with the NRG results of Fig. 3.

For clarity, the above discussion was confined to $H^*=$SEAM. However, it can be generalized straightforwardly to the non-symmetric case with $e_{\sigma e}^f \not= -\frac{1}{2}U$, as long as $H^*$ remains in the LM-regime, with $\bar{n}_e \approx 1$ (see Appendix 4). In the LM regime, the lineshape depends on $e_{\sigma e}^f$ and $U$ only through their influence on $T_K$, and hence $A^{\text{LM}}_{\sigma}(\nu)$ is a universal function of $\nu$ and $T_K$. This is illustrated in Fig. 4 for five lineshapes, shown in the lower left panel, corresponding to different choices of $e_{\sigma e}^f$ and hence different $T_K$-values (as indicated in inset). When these lineshapes are rescaled as $A_{\sigma}(\nu)/A_{\sigma}(T_K)$ vs. $\nu/T_K$ (main panel), they collapse onto a universal scaling curve within the LM regime $T_K \approx \nu \lesssim |\epsilon^f_{\sigma e}|$. An experimental observation of such a scaling collapse would be a smoking gun for the existence of Kondo correlations.
Small detuning dependence and the tunable Kondo-edge singularity – the non-perturbative regime. As \( \nu \) is lowered through the bottom of the LM regime, \( J(\nu) \) increases through unity into the strong coupling regime, and \( A_\nu(\nu) \) monotonically crosses over to SC behavior. In this limit, the assumption of a FR that remains unaltered by the interactions with the QD breaks down, requiring an alternative approach for determining \( A_\nu(\nu) \).

For small detuning, i.e. long times \( t > 1/T_K \), a screening cloud builds up that tends to screen the local moment into a spin singlet, as visible in the decay of \( \tilde{m}_c(t \to \infty) \to 0 \) (see Fig. 2). The screened spin singlet acts as a source of strong potential scattering for other FR electrons, causing the phase of each mode \( k\sigma \) to shift by \( \delta_\sigma(\varepsilon_{k\sigma}) \) relative to its value for \( H^1 \). This regime can be described by a strong-coupling fixed-point Hamiltonian \( H_{SC} + H_{SC} \) due to Nozières (given in Appendix 6). It is formulated purely in terms of these phase-shifted e-electrons and makes no reference to e-level operators at all, since in the SC-regime the local moment is fully screened. Thus, the fixed-point perturbation strategy used above cannot be applied here.

This hurdle can be overcome by working in the time-domain, and relating the correlator \( G_{ec}(t) \) mentioned after Eq. [5] to the X-ray edge problem. The latter deals with the absorption lineshape for an incident X-ray to suddenly dislodge an electron from an atomic core level, placing it in the conduction band and leaving behind a core hole (i.e. a local scattering potential). Our situation is similar, in that the screened singlet also serves as local scattering potential, but more complex, in that this potential is not turned on suddenly, but emerges only as the long-time limit of the dynamical build-up of a Kondo cloud to screen the local spin. Nevertheless, in the long-time limit an equation of motion approach can be used to relate the correlator \( G_{ec}(t) \) to a similar one, \( G_{ec}(t) \), which involves only conduction band electrons and whose form is known from the X-ray edge problem (see Methods and Appendix 6 for details). This readily leads to a power-law divergence characteristic of X-ray edge problems, with the infrared singularity exponent \( \eta_\sigma = 2\Delta n_{ec} - \sum_{\sigma'}(\Delta n'_{e\sigma'})^2 \),

\[
A_{SC}(\nu) \propto T_K^{-1}(\nu/T_K)^{-\eta_\sigma},
\]

where the infrared singularity exponent \( \eta_\sigma \) depends on the e-level’s change in average occupation, \( \Delta n_{e\sigma} = \tilde{n}_{e\sigma} - \tilde{n}_{e\sigma} \). Eq. [8] may be regarded as a generalized version of “Hopfield's rule of thumb” [26], which was established rigorously in Refs. [23, 27]. It has an instructive physical interpretation, based on rewriting it as \( \eta_\sigma = 1 - \sum_{\sigma'}(\Delta n'_{e\sigma'})^2 \), where \( \Delta n'_{e\sigma'} = \Delta n_{e\sigma} - \delta_{\sigma\sigma'} \) is the charge difference in level \( e\sigma' \) between the final ground state \( |\infty\rangle \) (at time \( t \to \infty \)) and the state \( |0^+\rangle \) the system finds itself in just after photo-excitation of a spin-\( \sigma \) electron (at \( t = 0^+ \)). The “1” in \( \eta_\sigma \) represents a \( \nu^{-1} \) power law divergence: it may be thought of as arising from a detuned, virtual transition into a narrow e-level situated at \( \nu = 0 \) (giving a Lorentzian detuning factor \( 1/\nu^2 \)), followed by the creation of particle-hole pairs (with phase space \( \nu \)) to carry off the excess energy \( \nu \), resulting in a lineshape scaling as \( \nu/\nu^2 = \nu^{-1} \). The \( \sum_{\sigma'}(\Delta n'_{e\sigma'})^2 \) contribution to \( \eta_\sigma \) reflects Anderson orthogonality [13]: since \( |\infty\rangle \) and \( |0^+\rangle \) have localized \( e\sigma' \)-charges that differ by \( \Delta n'_{e\sigma'} \), their Fermi reservoir electrons see different scattering potentials, implying [12] that their overlap scales with effective system size \( L \sim \nu^{-1} \) as \( \langle \infty | 0^+ \rangle \sim L^{-\sum_{\sigma'}(\Delta n'_{e\sigma'})^2} \).

For the X-ray edge problem, \( \Delta n_{e\sigma} \) and hence \( \eta_\sigma \) are fixed by material parameters (the strength of the core-hole potential). In contrast, in the present case they can be tuned experimentally by sweeping \( \varepsilon_{k\sigma} \) and \( \varepsilon_{e\sigma} \) with a gate voltage or magnetic field (see Figs. 5b and Figure S3). This tunability can be exploited to study unusual aspects of Anderson orthogonality physics that had hitherto been inaccessible. In particular, if the system is tuned such that \( \tilde{n}_{e\sigma} = 0 \) and \( \tilde{n}_{e\sigma} = 1 \), Eq. [5] can be expressed as \( \eta_\sigma = \frac{1}{2} + 2m_e^{e\sigma} - 2m_e^{e\sigma} \), where the final magnetization \( m_e^{e\sigma} = \frac{1}{2}(\tilde{n}_{e\sigma} + \tilde{n}_{e\sigma} - 1) \) is a universal function of \( g_\sigma B/T_K \). (At very large fields, however, a bulk Zeeman field, neglected above, will spoil universality, see Fig. 4: Universal in the LM-regime. Lower left panel: Five lineshapes, corresponding to five different choices of \( \varepsilon_{k\sigma} \), indicated by color-coded dashed lines in upper panel, and arrows in main panel. The inset also gives \( \tilde{n}_{e\sigma} \), \( \Delta n_{e\sigma} \), and \( T_K \) as function of \( \varepsilon_{k\sigma} \). Main panel: When appropriately rescaled, the lineshapes collapse onto a universal curve in the LM-regime \( T_K \lesssim \nu \lesssim \varepsilon_{e\sigma} \). In the SC-regime \( T \lesssim \nu \lesssim T_K \), the curves do not collapse, since their exponents \( \eta_\sigma \) depend, via \( \Delta n_{e\sigma} \), on \( \varepsilon_{e\sigma} \). NRG parameters: \( U = 0.1D \), \( U_{eh} = 1.25U \), \( \Gamma = 0.062U \), \( T = 1.6 \cdot 10^{-8}\Gamma \), \( B = 0 \), \( \Lambda = 1.8 \). Kept states: 1024, \( \alpha = 0.5 \).
of our analysis. A broadened peak, is one of the most striking predictions
will turn a near-threshold divergence in the lineshape to
a change of spin orientation of the photo-excited electron
and once \( \nu \) is accompanied by the emergence of an absorption peak
of its spin (see Appendix 7). The fact that for \( \nu = |g_e B| \), associated with a transition into the upper
Zeeman-split e-level, broadened by Korringa relaxation
(see Appendix 8.) Thus, the exponents \( \eta_\sigma(B) \) are universal functions of \( g_e B/T_K \), with simple limits for small and
large fields (see Fig. 5b):

\[
\eta_{\text{lower/upper}} \rightarrow \begin{cases} 
\frac{1}{2} & (|B| \ll T_K), \\
\pm 1 & (|B| \gg T_K).
\end{cases}
\]  

The notation \( \sigma = \text{ "lower" or "upper" } \) distinguishes whether the spin of the photo-excited electron (selectable
by choice of circular polarization of the incident light) matches the spin of the lower or upper of the Zeeman-
split e-levels, respectively. The sign difference between \( \eta_{\text{lower}} \) and \( \eta_{\text{upper}} \) for \( |B| \gg T_K \) arises since the change in
local charge becomes fully asymmetric, \( \Delta n_{\text{lower}} \rightarrow -1 \)
while \( \Delta n_{\text{upper}} \rightarrow 0 \); as a result, for \( \sigma = \text{ lower Anderson } \) orthogonality is completely absent \( (\Delta n_{\text{corr}} = 0) \), whereas
for \( \sigma = \text{ upper } \) it is maximal \( (\Delta n_{\text{corr}}' = 1) \). It follows, remarkably, that a magnetic field tunes the strength of Anderson orthogonality, implying a dramatic \( \sigma \)-dependence of the evolution of the lineshape \( A_\sigma(\nu) \sim \nu^{-\eta_\sigma} \) with increasing \( |B| \) (Fig. 5a): For \( A_{\text{lower}}(\nu) \), the near-threshold singularity becomes stronger, tending towards \( \nu^{-1} \). In contrast, for \( A_{\text{upper}}(\nu) \) the singularity becomes weaker, and once \( \eta_{\text{upper}} \) turns negative, changes to an increasingly strong power-law decay, tending toward \( \nu^1 \); this is accompanied by the emergence of an absorption peak
near \( \nu = |g_e B| \), associated with a transition into the upper Zeeman-split level, broadened by Korringa relaxation
of its spin (see Appendix 7). The fact that for \( |B| > T_K \) a change of spin orientation of the photo-excited electron
will turn a near-threshold divergence in the lineshape to
a suppression that constitutes the low-frequency side of a broadened peak, is one of the most striking predictions of our analysis.

**Magnetic field dependence-dependence of the absorption threshold.** The shift of the absorption
threshold frequency \( \omega_{\text{th}}(B) = E_0^G - E_0^F \) with magnetic field can be written as
\( \omega_{\text{th}}(B) = \omega_{\text{th}}(0) + \frac{1}{2} \frac{\partial g_e B}{\partial B} \). The first term reflects the Zeeman energy of the photo-
excited hole (which has pseudo-spin 1/2), the second the \( B \)-dependence of the ground-state energy of the electron system. The general \( T = 0 \) relation \( g_e m_e^a = \partial E_0^G/\partial B \) implies that the differential threshold shift offers a direct way of experimentally measuring the local moment difference between the final and initial ground states:

\[
\delta \omega_{\text{th}} = \frac{\partial \omega_{\text{th}}(B)}{\partial B} = g_e |m_e^a(B) - m_e^b(B)|. 
\]  

Also, for \( \bar{n}_e \approx 0 \) and \( n_e \approx 1 \), where Eq. (9) applies, this quantity can be related to the infrared singularity exponents, allowing for a consistency check. Moreover, the asymptotic behavior of \( m_e^a \) for small fields \( (m_e^a = -g_e B \chi_0, \chi_0 = 1/4T_K \) is the static susceptibility) and large field \( (|m_e^a| = \frac{1}{2}) \) implies:

\[
\delta \omega_{\text{th}} = \begin{array}{ll}
\frac{-(g_e B)^2}{8T_K} & (|B| \ll T_K), \\
\frac{-g_e B}{2} & (T_K \ll B \ll |\varepsilon_c|) 
\end{array}
\]  

We note that a formula that interpolates through both of these regimes is given by \( \delta \omega^\star_{\text{th}} = T_K - \frac{1}{2} \sqrt{(g_e B)^2 + (2T_K^2) \omega_{\text{th}}(0)} \) which, up to numerical pre-factors, has a similar functional form as the expression for
the ground state energy of the s-d model derived in Ref. [23]. The quadratic \( B \)-dependence of \( \omega^\star_{\text{th}} \) for small fields offers a straightforward way to determine the Kondo temperature experimentally. The accessibility of the \( B \)-dependence of the ground state energy and e-level magnetization via the absorption threshold is a remarkable advantage of the proposed optical probe of Kondo physics in this paper – these quantities are not accessible via transport measurements.
We have demonstrated that optical absorption in a single quantum dot can be used to implement a quantum quench - a sudden change in the Hamiltonian governing the dynamics of the many-body system. Given that the relevant quantum dot parameters are tunable via external electric and magnetic fields, this system constitutes one of the rare, if not unique, experimentally accessible solid-state systems where a tunable quantum quench could be realized. Our work sets the stage for exploring numerous further interesting problems, such as (i) the effect of an exchange interaction between e- and h-levels; (ii) the effect of nuclear spins on the electron spin dynamics; (iii) using a pump-probe protocol to study the non-equilibrium time evolution even more directly; (iv) studying the coherence of optical Raman transitions, where the virtually excited intermediate state is a Kondo correlated state of the QD electron and an adjacent fermionic reservoir, and (v) attempting to exploit strong correlations to realize a “Fermionic quantum bus” between two distant QD spins. The presented results have an important bearing on the quantum control of a spin degree of freedom in nano-structures and on quantum optical techniques used to study reservoirs composed of fermionic constituents.

Methods

A. Numerical Renormalization Group for calculation of optical absorption lineshape

The optical absorption lineshape given by Fermi’s golden rule Eq. 2, can be calculated at finite temperatures using full density matrix (FDM) numerical numerical renormalization group (NRG) \[14\]. Because Eq. 2 contains matrix elements between initial and final eigenstates of different Hamiltonians, \( H^I \) and \( H^F \), two separate NRG runs (NRG run \#1 and \#2) are required to calculate the initial and final eigenstates (\( \{|m_i\} \) and \( \{|n_i\} \)) as well as eigenenergies (\( \{E^I_{m_i}\} \) and \( \{E^F_{n_i}\} \)). The double sum in Eq. 2, over all initial and final eigenstates, is performed via a “backwards” run from the end to the beginning of the Wilson chain\[14\]: for each shell \( k \), the contribution towards the initial density matrix \( \hat{\rho}^I \) from that shell (obtained using data from NRG run \#1), and the matrix elements \( \langle n|e_{\sigma}^I|m_i\rangle^2 \) between shell-\( k \) eigenstates from NRG runs \#2 and NRG \#1 are calculated, and binned according to the corresponding frequency difference \( E^F_{n_i} - E^I_{m_i} \). See Appendix 2 for further details.

B. Non-equilibrium dynamics via NRG

The expectation value of an observable \( \hat{B} \) after absorption is given by \( \hat{B}(t) = \text{Tr} \left( \hat{\rho}^I(t)\hat{B} \right) \) where the time evolution is governed by the final Hamiltonian, \( \hat{\rho}^I_p(t) \equiv e^{-\hat{H}^I t}\hat{\rho}_p e^{\hat{H}^I t} \). For reasons discussed in Appendix 3 we find it convenient to take \( \hat{\rho}^I_p = \hat{\rho}_p/[1 - \hat{n}_{\sigma}^I] \). The Fourier transform of \( \tilde{B}(t) \), \( \tilde{B}^e(\omega) = \int dt e^{i\omega t}\tilde{B}(t) \), can be expressed in Lehmann representation:

\[
\tilde{B}^e(\omega) = \sum_{n,n'} \langle n'|\hat{\rho}_{n'}|n\rangle\langle n|\hat{B}|n'\rangle \cdot 2\pi\delta(\omega - E^I_{n'} + E^I_{n}) \tag{11}
\]

This expression can be calculated using FDM-NRG. See Appendix 3 for details.

C. Fixed-point perturbation theory

To calculate \( A^e(\nu) \) at \( T = 0 \) for \( \nu \gtrsim T_K \), set \( H^F \to H^F_0 \) and expand in powers of \( H^F_0 \), for \( r = FO \) or LM. At zeroth order this yields a peak \( A^e(\nu) \sim \delta(\nu) \), which is irrelevant for \( \nu \gtrsim T_K \). To lowest non-vanishing order in \( H^F_0 \)

\[
A^e(\nu) = -\frac{2}{V^2} \text{Im} \langle i(G|e_{\sigma}H^F_0 - \frac{1}{\nu}H^F_0 e_{\sigma}|G) \rangle \tag{12}
\]

where we shall evaluate for \( B = 0 \), \( |e_{\sigma}^I,\nu\rangle = \frac{1}{2} |U \) and \( |G\rangle \equiv \prod_{k_{\sigma}, \epsilon_k} c^\dagger_{k\sigma} |\text{Vac}\rangle \). We find

\[
A^e(\nu) = -\frac{2}{V^2} \text{Im} \left[ T^e(\nu) \right] \tag{13}
\]

where \( T^e(\nu) = 2(1/\pi \rho) F^e_{\nu} \{ -i\theta(t) \langle i(G|e_{\sigma}(t)\tilde{e}_{\sigma}(\nu)\rangle |G\rangle \} \) in the FO-regime \( (F^e_{\nu} \) is the Fourier-transform operator \( F^e_{\nu} \{ A \} \equiv \int dt e^{i\nu(\nu + \epsilon_m)}d_{m\nu}(t) \). In this regime, the absorption process can be understood as a two-step process consisting of a virtual excitation of the QD resonance, followed by a tunneling event to a final free-electron state above the Fermi-level. Therefore, the absorption rate is proportional to the tunneling density of states (into the FR) at detuning \( \nu \). On the other hand, in the LM regime, \( T^e(\nu) = 3/8 (J/\rho)^2 F^e_{\nu} \{ -i\theta(t) \langle i(G|\tilde{e}_{\sigma}(\nu)\epsilon_{\sigma}(\nu)\rangle |G\rangle \} \) where we used \( \langle \tilde{e}_{\sigma}^e(t)\tilde{e}_{\sigma}(\nu)\rangle = \langle \tilde{e}_{\sigma}^e(\nu)\tilde{e}_{\sigma}(\nu)\rangle /2 \). This result reiterates that the intermediate detuning probes spin fluctuations, as observed in the dynamics (Fig. 2). Evaluating these correlation functions yields Eq. 5 and Eq. 9. In Eq. 10, we have inserted the scale-dependent exchange interaction \( J(\nu) = \ln^{-1}(\nu/T_K) \) which results from the logarithmic enhancement of the exchange interaction within perturbative RG. The finite-temperature form \( (T \gg T_K) \) of the lineshape is discussed in Appendix 5 (see also Fig. S2).

D. Absorption in the strong-coupling regime and Fermi-edge physics

Eq. 9 can be expressed as the Fourier transform, \( G^{ee}(\nu) = F^e_{\nu} \{ G^{ee}(t) \} \), of a correlator

\[
G^{ee}(t) = -i\theta(t) \langle \tilde{e}_{\sigma}(t)\tilde{e}^\dagger_{\sigma}(0) \rangle \tag{14}
\]

involving operators defined to have an anomalous time dependence, \( \tilde{O}(t) = e^{iH^I t}\tilde{O} e^{-iH^I t} \). This anomalous time
dependence, involving both \(H^i\) and \(H^f\), reflects the fact that the creation of a hole during optical absorption abruptly lowers the e-level. We next relate \(\mathcal{G}_{cc}^{\sigma}(\nu)\) to a similarly-defined correlator of FR electrons,

\[
\mathcal{G}_{kk'}^{\sigma}(\nu) = \mathcal{F}_\nu \left\{ -i\theta(t) \langle c_{k\sigma}(t)c_{k'\sigma}^\dagger(t) \rangle \right\}
\]

(15)

using equations of motion in the asymptotic limit \(\nu \to 0\):

\[
\mathcal{G}_{kk'}^{\sigma}(\nu) \sim \frac{\nu^2 \mathcal{G}_{cc}^{\sigma}(\nu)}{(\nu_+ + \Delta - \varepsilon_{k\sigma})(\nu_+ - \Delta - \varepsilon_{k'\sigma})},
\]

(16)

where \(\Delta = \omega_\text{th} - \varepsilon_{h\sigma}\). This asymptotic relation is established in Appendix 6 and implies

\[
A_{\sigma}^{\text{sc}}(\nu) \sim \frac{2}{\pi \rho \Gamma} \text{Im} \mathcal{G}_{cc}^{\sigma}(\nu).
\]

(17)

where \(\mathcal{G}_{cc}^{\sigma}(\nu) = \sum_{kk'} \mathcal{G}_{kk'}^{\sigma}(\nu)\). To calculate \(\mathcal{G}_{cc}^{\sigma}(t)\) for \(t \gg 1/T_K\), we may now replace \(H^i \to H_e\) and \(H^f \to H_{\text{SC}} + H_{\text{SC}}^c\) in Eq. (17):

\[
\mathcal{G}_{cc}^{\sigma}(t) \sim i\langle G|e^{iH_e t}c_\sigma e^{-iH_{\text{SC}}^c}c_\sigma^\dagger|G\rangle_i.
\]

(18)

This response function is similar to that calculated in the X-ray edge problem ([22, 23, 24] and its calculation is standard (e.g. [24]) and yields (we show only the leading power law)

\[
\mathcal{G}_{cc}^{\sigma}(t) \sim t^{-|\delta_\sigma - \pi|^2 + \delta_\sigma^2/\pi^2}.
\]

(19)

where \(\delta_\sigma = \delta_\sigma(0)\) denotes the phase shifts at the Fermi energy. The phase shifts at the Fermi energy, in turn, are given by \(\delta_\sigma = \pi \Delta n_{\sigma}\), according to the Friedel sum rule ([24]), valid for \(T = 0\) and for arbitrary values of \(B\), \(n_e^c\) and \(n_e^c\). Collecting results, we find Eq. (17) and Eq. (18). See Appendix 6 for details.

Acknowledgments

AI and HET acknowledge support from the Swiss NSF under Grant No. 200021-121757. HET acknowledges support from the Swiss NSF under Grant No. PP00P2-123519/1. BB acknowledges support from the Swiss NSF and NCCR Nanoscience (Basel). JvD acknowledges support from the DFG (SFB631, SFB-TR12, De730/3-2, De730/4-1), the Cluster of Excellence Nanosystems Initiative Munich and in part the National Science Foundation under Grant No. NSF PHY05-51164. AI acknowledges support from an ERC Advanced Investigator Grant, and LG from NSF Grant No. DMR-0754613.

APPENDIX 1: THE MODEL AND ITS EXPERIMENTAL REALIZATION

We focus here on differential transmission spectroscopy of single semiconductor QDs in gated heterostructures with an adjacent tunnel-coupled electron reservoir. A possible realization is a Schottky diode structure ([14] where a voltage applied between a top Schottky contact and a two-dimensional electron gas (2DEG) in a modulation-doped GaAs layer underneath a layer of self-assembled InAs QDs, is used to adjust the relative energy of the QD electron with respect to the Fermi-energy of the 2DEG (Figure [21]). To realize the situation in Fig. 1 we consider here a range of gate voltages for which the QD is uncharged before the optical excitation (See Fig. 4, upper inset). In contrast to most of the earlier quantum optics experiments where the separation between the electron gas and the QD layer was kept large to ensure weak spin-flip co-tunneling, we focus here on structures with a small barrier where tunnel coupling of a QD electron is only a factor of 10 weaker than its single-electron charging energy. We estimate that with a 15 nm neutral GaAs barrier, the strength of the exchange interactions would be strong enough to yield Kondo temperatures exceeding 100 mK.

Spin-orbit interaction in InGaAs QDs leads to a splitting of about 200 meV between the \(J = 1/2\) and \(J = 3/2\) valence band states. In the limit of asymmetric quantum confinement typical for our QDs, the four \(J = 3/2\) bands further split into two Kramers doublets. The states with \(J_z = \pm 3/2\) along the growth (=strong confinement) direction have the lowest zero-point energy due to their heavy mass for motion along the growth direction and hence are the ones relevant for lowest energy optical excitations. The confinement+strain induced splitting between the \(J_z = \pm 3/2\) (heavy-hole) and \(J_z = \pm 1/2\) (light-hole) states is estimated to be around 20 meV. In the main text, the spin label of the valence band electrons (or holes, \(h_\sigma\)) therefore refer to pseudo-spin \(J_z = \pm 3/2\), and the hole Zeeman energy has the form \(\sigma 3/2 g_B B\).

This situation leads to near perfect correlation between the circular polarization of the excitation light and the spin of the optically excited electron in InGaAs QDs (as well as any QD structure with possibly the exception of chemically synthesized nanocrystals). The hole g-factor in these QDs is highly anisotropic: along the growth direction, the value is around 1.2. In plane g-factor ranges from 0 – 0.5.

For the calculations in the main body of the paper, we assume the electron-hole recombination rate (typically of order \(\Gamma_{\text{eh}} \approx 1 \mu\text{eV}\)) to be negligible compared to all other scales, take the hole level to lie within the gap of the FR and neglect its coupling to the latter. The newly-created electron-hole pair will experience a mutual Coulomb attraction \(U_{\text{eh}}(>0)\) and a much weaker exchange interaction \(J_{\text{eh}}\) (typically of order \(J_{\text{eh}} \approx 200 \mu\text{eV}\)) ([12]).

While the interplay of the intra-dot electron-hole exchange interaction and the Kondo correlations is interesting on its own right, the effects of electron-hole exchange can experimentally be avoided in two ways. One possible alternative is to start out with a single-hole charged QD; the experiments on single QDs embedded in n-type Schottky structures have already shown that the life-
time of such an optically charged state could well exceed 100 μsec. Resonant optical excitation of such a single-hole charged QD leads to the formation of the so-called $X^{1+}$ trion which consists of two valence-band heavy-holes forming a singlet and a single e-level electron with vanishing electron-hole-exchange interaction. Another possible alternative is using indirect excitons in coupled QDs where the electron and the hole wave-functions have vanishing spatial overlap.

Here, we assume $J_{eh} \ll T_K$, or an experimental realization where it can be neglected, so that the hole has no dynamics after creation. With the hole “frozen”, the electron dynamics can be described by a pure Anderson model (AM), Eq. (1), for the e-level (involving only $\varepsilon_\sigma$, not $h_\sigma$), whose initial and final energies before and after absorption, $\varepsilon_\sigma^m$ ($a = i, f$), are related by $\varepsilon_\sigma^m = \varepsilon_\sigma - \delta_{af} U_{eh}$ (Fig. 1b). The downward pull of the Coulomb attraction $U_{eh}$ stabilizes the excited electron against decay into the FR, provided that $\varepsilon_\sigma^f$ lies below the FR’s Fermi energy $\varepsilon_F = 0$.

**APPENDIX 2: NUMERICAL RENORMALIZATION GROUP AND FERMI’S GOLDEN RULE**

The numerical renormalization group (NRG) is an iterative method for numerically diagonalizing quantum impurity models such as Anderson impurity Hamiltonians $H^a$ ($a = i, f$) specified around Eq. (1). The spectrum of states of the Fermi reservoir is coarse-grained using a logarithmic discretization scheme governed by a parameter $\Lambda > 1$ (typically $\Lambda = 2$), followed by an exact mapping of the discretized model onto a semi-infinite chain, the so-called Wilson chain, whose hopping amplitudes decay exponentially along the chain, as $t_k \sim \Lambda^{-k/2}$. This produces a separation of energy scales and makes it possible to diagonalize the Hamiltonian iteratively: knowing the eigenstates of a chain of length $k - 1$, one adds site $k$ and calculates the “shell” of eigenenergies of the Hamiltonian for the chain of length $k$. The high-lying eigenstates of that shell are “discarded”, while the low-lying states are “kept” and used for the next iteration. The spectrum of eigenenergies so obtained typically flows past one or more non-stable fixed-points and finally converges towards a stable fixed point, whereupon the iterative procedure can be stopped. In practice one thus deals with a finite Wilson chain, whose length is set by the smallest energy scale in the system (e.g. the Kondo temperature, temperature, or magnetic field). By combining NRG data from all iterations, it is possible to construct a complete set of approximate many-body eigenstates of the full Hamiltonian. These can be used to evaluate equilibrium spectral functions via their Lehmann-representations; at finite temperatures, this can be done using the full density matrix (FDM)-NRG.

Since Eq. (2) expresses the Fermi golden rule absorption rate via a Lehmann representation, it, too, can be evaluated using NRG. However, it contains matrix elements between initial and final states that are eigenstates of different Hamiltonians, $H^1$ and $H^f$. Hence, two separate NRG runs are required to calculate these (similar in spirit to what is done for time-dependent NRG). The strategy is then as follows:

- **NRG run #1** generates a complete set of approximate eigenstates $|m\rangle_i$ and eigenenergies $E_{m}^i$ for the initial Hamiltonian $H^1$ (without exciton).
- **NRG run #2** generates a complete set of approximate eigenstates $|n\rangle_f$ and eigenenergies $E_{m}^f$ for the final Hamiltonian $H^f$ (with exciton).
- The double sum in Eq. (2), over all initial and final eigenstates, is performed via a “backwards” run, with site index $k$ running from the end to the beginning of the Wilson chain: for each shell $k$, the contribution towards the initial density matrix $\rho_i$ from that shell (obtained using data from NRG run #1), and the matrix elements $|\langle n|e_{\sigma}^f|m\rangle|^2$ between shell-$k$ eigenstates from NRG runs #2 and NRG #1 are calculated, and binned (see below) according to the corresponding frequency difference $E_{m}^f - E_{m}^i$.
- The $T = 0$ threshold frequency for the onset of absorption is given by the difference of groundstate energies of NRG runs #2 and #1, $\omega_{th} \equiv E_{G}^f - E_{G}^i$. The absorption spectrum is expected to have divergences at the threshold $\omega_{th}$, hence all frequency data are shifted by the overall threshold energy $\omega_{th}$ prior to binning. (For finite temperature, the sharp onset is broadened and divergencies are cut off.)
- The discrete eigenenergies of shell $k$ are spread over an energy range comparable to the characteristic energy $\Lambda^{-k/2}$ scale of that iteration, which decreases exponentially with $k$. Thus, the bins used for collecting the discrete data are likewise chosen to have widths decreasing exponentially with decreasing energy. The discrete, binned data are subsequently broadened using a log-Gaussian broadening scheme, characterized by a broadening parameter $\alpha$, typically taken as $\alpha = 0.6$. For finite temperature, the maximum of the absorption

FIG. S1: The Schottky diode structure for the experimental observation of the optical signatures of the Kondo effect.
peak occurs at a frequency $\nu_{\text{max}}$, that is slightly larger than the $T = 0$ threshold frequency at $\nu = 0$ (see Fig. 3a). Thus, for finite temperature, the binning procedures (both the binning frequencies and the frequency ranges in which we change from log-Gaussian to Gaussian) for frequencies above and below $\nu_{\text{max}}$ were set up to be symmetric with respect to $\nu_{\text{max}}$. This ensures that the $A_\nu(\nu)$ curves for $\nu$ smaller and larger than $\nu_{\text{max}}$ match smoothly.

**APPENDIX 3: TIME EVOLUTION VIA NRG**

The nonequilibrium time evolution of the system in response to the sudden creation of an \( e^i \hbar \sigma \) exciton at time \( t = 0 \) can be calculated using time-dependent NRG\[10\].

The sudden addition of a $\sigma$-electron in the e-level projects the system’s initial density matrix $\hat{\rho}_i$ onto a projected density matrix $\hat{\rho}_p$,

$$\hat{\rho}_p = \sum_m |m\rangle_i \langle m| \rightarrow \hat{\rho}_p = e_\sigma^\dagger \hat{\rho} e_\sigma . \quad (S1)$$

Its norm is smaller than 1,

$$\text{Tr} \hat{\rho}_p = \text{Tr} \left( \hat{\rho} e_\sigma e_\sigma^\dagger \right) = 1 - n_{\sigma \sigma}^i , \quad (S2)$$

reflecting the fact that states in which the e-level had already contained a $\sigma$-electron prior to absorption are projected to 0. \( n_{\sigma \sigma}^i = (\bar{n}_{\sigma \sigma})_i \) denotes an expectation value w.r.t. to \( H^i \). After this projection, the e-level occupancies for spin $\sigma'$ are reset (or “reinitialized”) to be

$$\text{Tr} (\hat{\rho}_p \hat{n}_{\sigma'\sigma'}), \quad \left( \begin{array}{c} \bar{n}_{\sigma \sigma}^i = 1 - \bar{n}_{\sigma' \sigma'}^i = 1 - \bar{n}_{\sigma' \sigma'}^i \quad \text{for} \quad \sigma' = \sigma \\ \bar{n}_{\sigma \sigma}^i \quad \text{for} \quad \sigma' = \bar{\sigma} \end{array} \right) \quad (S3)$$

where $\bar{n}_{\sigma \sigma}^i$ and $\bar{n}_{\sigma' \sigma'}^i$ are the initial probabilities (with respect to \( H^i \)) for the e-level to have been completely empty (00), or to have contained no $\sigma$-electron but an $\bar{\sigma}$-electron (0$\bar{\sigma}$), respectively.

For the sake of studying the time evolution after absorption, we find it convenient to take $\hat{\rho}_p = \hat{\rho}_p^i / [1 - \bar{n}_{\sigma \sigma}^i]$ as starting density matrix, normalized such that $\text{Tr} \hat{\rho}_p^i = 1$ and $\text{Tr} \hat{\rho}_p^i \bar{n}_{\sigma \sigma} = 1$. Its subsequent time evolution is governed by the final Hamiltonian, $\hat{\rho}_p(t) = e^{-iH^f t} \hat{\rho}_p^i e^{iH^f t}$.

The corresponding expectation value of an observable $\hat{B}$ is thus given by

$$\hat{B}(t) = \text{Tr} \left( \hat{\rho}_p^i(t) \hat{B} \right) . \quad (S4)$$

Fourier-transformed to frequency space using $\hat{B}(\omega) = \int dt \ e^{i\omega t} \hat{B}(t)$, one obtains

$$\hat{B}(\omega) = \sum_{n,n'} \langle n| \hat{\rho}_p^i|n'\rangle_{1f}|n|\hat{B}|n'\rangle_{1f} \cdot 2\pi \delta (\omega - E_{n'} + E_{n}') \quad (S5)$$

The latter expression, being written in Lehmann representation, is again well-suited for FDM-NRG\[10\]. Following \[10\], it is again possible to formulate the procedure such that matrix elements are always calculated within the same Wilson shell.

The discrete data contributing to $\hat{B}(\omega)$ is binned and smoothened using a log-Gaussian broadening function, as described in the previous section, but using a smaller broadening parameter, $\alpha = 0.3$.

**APPENDIX 4: PERTURBATION THEORY FOR THE EXCITONIC ANDERSON MODEL**

The optical absorption lineshape for a $\sigma = +$ exciton and the excitonic Anderson Model can be calculated via a perturbation expansion up to second order in the tunnel coupling $v = \sqrt{1/\pi \rho}$ (we keep here the momentum dependence of $v_k$) for the initial and final states entering Eq. (2). (The wavefunctions used below are similar in structure to those used in Ref.\[10\] for variational calculations of various emission and absorption spectra of Ce compounds.)

We define the empty e-level, h-level and unperturbed FR as $|0\rangle = |0\rangle_e \otimes |0\rangle_h \otimes |0\rangle_F$. The bare excitonic resonance frequency is $\omega_0 = \omega_0 + U_{ch} + \epsilon_h$ which also marks the approximate onset of the edge (threshold) behavior. We write the initial state $|G\rangle_i$ expanded to second order in $v_k$ as

$$|G\rangle_i = |1 + \sum_{k < 0} \frac{v_k}{\epsilon_k - \epsilon_e} \hat{c}_{k,\sigma}^\dagger \hat{c}_{k,\sigma} |0\rangle + \sum_{k < 0} \frac{v_k v_{k'}}{\epsilon_k - \epsilon_{k'}} |0\rangle \quad (S6)$$

The set of final states are defined by particle number conservation. A few possible zeroth order final states accessible from the initial state (to low orders in $v_k$) are:

$\epsilon_{k,\sigma} \hat{c}_{k,\sigma}^\dagger |0\rangle (X^+ \text{ excitation}), \epsilon_{k,\sigma} \hat{c}_{k,\sigma}^\dagger \hat{c}_{k',\sigma}^\dagger |0\rangle (X^- \text{ excitation})$ and $\epsilon_{k,\sigma}^\dagger \epsilon_{k',\sigma} \hat{c}_{\bar{k},\bar{\sigma}}^\dagger \hat{c}_{\bar{k},\bar{\sigma}}^\dagger |0\rangle (X^0 + e-h \text{ excitation}).$ Note that the last class of states includes for $k = k'$ the bare neutral
For the asymmetric Anderson model, this threshold splits excitations in the FR. The final state energies to lowest order in e-h exciton final state. The relevant final states are then

\[ |X^+_{kσ}| = \left[ 1 + \frac{v_k}{ε_k - ε_e + U|c_σ^+c_σ|} \right] + \sum_{k'} \frac{v_{k'}}{ε_{k'} - ε_e + U|c_σ^+c_{σ'}|} |c_{k',σ}^+h_0^{|0}\rangle \]  

\[ |X^-_{kσ}| = \left[ 1 + \frac{v_k}{ε_k - U - ε_e - ε_{k,σ}^+} \right] + \sum_{k'} \frac{v_{k'}}{ε_{k'} - U - ε_e - ε_{k',σ}^+} |c_{k',σ}^+h_0^{|0}\rangle \]  

\[ |X^0_{kσk'σ'}(μ)| = \left[ 1 + \delta_{σ',μ} \frac{v_k}{ε_k - U + U|c_σ^+c_σ|} \right] + \sum_{k'} \frac{v_{k'}}{ε_{k'} - U + U|c_σ^+c_{σ'}|} |c_{k',σ}^+h_0^{|0}\rangle \]  

We have included the lowest order contributions in e-h excitations in the FR. The final state energies to lowest order in \( v_k \) are given by

\[ E_0^{(X^+)} = ω_{ch} + [ε_k - (ε_e - U|c_σ^+c_σ|)], \]  

\[ E_0^{(X^-)} = ω_{ch} + [(ε_e - U|c_σ^+c_σ|) - ε_k], \]  

\[ E_0^{(X^0)} = ω_{ch} + ε_e - U|c_σ^+c_σ| \]  

For SEAM, the analytical form of the lineshape is:

\[ A_T(ν) = \frac{3π}{4} \left( \frac{π}{ν} U \right) \left[ \frac{2}{U^2 + ν} + \frac{5}{U^2 - ν} \right] + \frac{1}{ν^3} \log \left[ \frac{(U^2 + ν)}{U^2 - ν} \right] + \frac{1}{ν^2} \log \left[ \frac{U + ν}{U^2 - ν} \right] + \frac{10Γ}{ν^2} \cdot θ(ν - U/2) \]  

The singularity at \( ν = U/2 \) can be related to a tunneling-assisted \( X^- \) resonance. The intermediate state there is an \( X^- \) charging state which is reached from the \( X^0 \) state by a FR electron tunneling in, and thereby satisfying energy conservation. We expect that a calculation including higher orders of perturbation theory will regularize this singularity.

The FO “threshold” term \( θ(ν - U/2) \), which corresponds to Eq. (5), is specific to the symmetric Anderson model. For the asymmetric Anderson model, this threshold splits into two, at \( ν = |ε_{ec}| \) and \( ν = ε_{eo}^+ + U \).

**FIG. S2:** The absorption lineshape at high temperatures. For \( T \gg T_K \), the log-log plot shows the two portions of the absorption lineshape for red-detuning (\( ν < ν_{max} \)) and blue-detuning (\( ν > ν_{max} \)) with respect to the frequency \( ν_{max} \) (at which \( A_T(ν) \) reaches its maximum). Here, \( γ_{Kor} = 0.15 T \). NRG parameters: \( U = 0.1D, ε_σ^+ = 0.75U, ε_e = -0.5U, \) \( Γ = 0.03U, T_K = 5.9 \cdot 10^{-6} Γ, T = 100 T_K, B = 0, \) \( Λ = 1.8, \) Kept states: 1024, \( α = 0.5 \).

**APPENDIX 5: FINITE TEMPERATURE FIXED-POINT PERTURBATION THEORY**

To calculate \( A_T(ν) \) for \( T \gg T_K \), we set \( H^r \rightarrow H^r + H^r \) in Eq. (3) and expand in powers of \( H^r \), for \( r = FO \) or LM. Note that the proper finite-T generalization of Eq. (3) is \( G_σ^r(ν) = \mathcal{F}_ν \{ -iθ(t) (ε_σ(t))_i \} \). Let us focus only on the temperature dependence of the lineshape for \( |ν| < |ε_{ec}| \) and \( T \gg T_K \):

\[ A_T(ν) = \frac{3π}{4} \frac{γ_{Kor}(ν, T)}{1 - e^{-ν/T}} U^2 + \frac{γ_{Kor}^2(ν, T)}{3} \]  

where \( γ_{Kor}(ν, T) \) is the scale-dependent Korringa relaxation rate given by

\[ γ_{Kor}(ν, T) = \begin{cases} \frac{πT}{ln^2 |T/T_K|} & \text{for } |ν| < T_K, \\ \frac{πν}{ln^2 |ν/T_K|} & \text{for } |ν| > T_K \end{cases} \]  

This expression is compared to the lineshape calculated by NRG in Fig. S2. Note that for \( ν > T_K \), we recover Eq. (6).

**APPENDIX 6: EVALUATION OF THE ABSORPTION LINESHAPE IN THE STRONG-COUPLING REGIME**

As we descend to detunings well below the Kondo scale, \( ν < T_K \), the physics is governed by the strong coupling fixed point. It describes a fully screened singlet, acting as source of strong potential scattering for other FR electrons, causing the phase of each mode \( kσ \) to shift by \( δσ(ε_{kσ}) \) relative to its value for \( H^r \). According
to Nozières[21] (see also [3]), the fixed point Hamiltonian at \( B = 0 \) is \( H_{SC} = \sum_{k} \varepsilon_{k} c_{k\sigma}^{\dagger} c_{k\sigma} \), where tildes denote operators representing phase-shifted modes. The leading relevant perturbation for \( 0 < B \ll T_{K} \) has the form[21]

\[
H_{SC} = \sum_{k} \sigma_{g} B \left[ \frac{\theta_{k\sigma}}{\pi \rho_{g}} + \rho_{\sigma} / (\pi \rho_{K}) \right].
\]

We shall not explicitly use this fixed point Hamiltonian, however, since the strategy (described in Methods section) of perturbing around the fixed point is of no use for calculating \( G_{cc}^{\sigma}(\nu) \) of Eq. (3). The reason is that \( G_{cc}^{\sigma}(\nu) \) is formulated in terms of \( c_{\sigma} \) and \( c_{\sigma}^{\dagger} \) operators, whose dynamics is determined by higher-energy excitations of the FR not described by \( H_{SC} + H_{SC} \). To circumvent this problem, we use an equation of motion approach to first derive an asymptotic relation between the impurity Green’s function and the Green’s function of the itinerant electrons.

We start by noting that the correlator occurring in Eq. (3) can be expressed as the Fourier transform, \( G_{cc}^{\sigma}(\nu) = \mathcal{F}_{\nu} \{ G_{cc}(t) \} \), of a correlator

\[
G_{cc}^{\sigma}(t) = -i\partial_{t} \langle \psi_{\sigma}(t) \psi_{\sigma}^{\dagger}(0) \rangle,
\]

involving operators defined to have an anomalous time dependence, \( \tilde{O}(t) = e^{iH_{f} t} \tilde{O} e^{-iH_{f} t} \). This anomalous time dependence, involving both \( H_{f} \) and \( H_{K} \), reflects the fact that the creation of a hole during optical absorption abruptly lowers the e-level.

To relate \( G_{cc}^{\sigma}(\nu) \) to a similarly-defined correlator \( G_{cc}^{\sigma}(\nu) \), we note that the anomalous time-dependence of \( c_{\sigma} \) implies the equation of motion

\[
i\partial_{t} c_{k\sigma}^{\dagger}(t) = [\varepsilon_{k\sigma}(t), H_{f}^{\dagger} + c_{k\sigma}(t)(\varepsilon_{k\sigma} - U_{\text{ch}} n_{e}) \rangle,
\]

where \( \nu = \sqrt{\nu^{2} / \pi \rho} \). Inserting this into the definition of \( G_{cc}^{\sigma}(t) \) one finds

\[
i\partial_{t} G_{kk}^{\sigma}(t) \sim [\varepsilon_{k\sigma}^{\dagger}(t), H_{f}^{\dagger} + c_{k\sigma}(t)(\varepsilon_{k\sigma} - U_{\text{ch}} n_{e})] + v G_{cc}^{\sigma}(t),
\]

where terms that become subleading for \( t \to \infty \) have been dropped (a term containing \( \tilde{O}(t) \); and one containing \( \langle c_{k\sigma}(t) n_{e} c_{k\sigma}^{\dagger}(0) \rangle \); which contains more operators and hence decays more quickly with time than the correlators that were retained). Similarly one finds

\[
i\partial_{t} G_{kk}^{\sigma}(t) \sim [\varepsilon_{k\sigma}^{\dagger}(t), H_{f}^{\dagger} + c_{k\sigma}(t)(\varepsilon_{k\sigma} - U_{\text{ch}} n_{e})] + v G_{cc}^{\sigma}(t),
\]

where the cyclic property of the trace was used to write

\[
\langle \psi_{\sigma}(t), H_{f}^{\dagger} c_{k\sigma}^{\dagger}(0) \rangle = \langle \psi_{\sigma}(t), [H_{f}^{\dagger}, c_{k\sigma}^{\dagger}] \rangle.
\]

Fourier-transforming Eqs. (S20) and (S19) using the convention stated just before (S19) and eliminating \( G_{cc}^{\sigma}(\nu) \), we readily find the asymptotic relation:

\[
G_{cc}^{\sigma}(\nu) = \sum_{kk'} G_{kk'}^{\sigma}(\nu) - \pi \rho \Im G_{cc}^{\sigma}(\nu),
\]

where the double sum \( \sum_{kk'} = \rho^{2} \int d\varepsilon_{k} d\varepsilon_{k'} \) is exhausted by two \( \delta \)-functions, since \( |\Delta| \) is of order \( |\varepsilon_{f}| \), which we assume to be smaller than the bandwidth \( D \). Eq. (S23) implies

\[
A_{\sigma}^{SC}(\nu) \sim \frac{2}{\pi \rho} \Im G_{cc}^{\sigma}(\nu).
\]

To calculate \( G_{cc}^{\sigma}(t) \) for \( t \gg 1 / T_{K} \), we may now replace \( H' \to H_{e} \) and \( H_{f} \to H_{SC} + H_{SC} \) in Eq. (S24):

\[
G_{cc}^{\sigma}(t) \sim i \langle G | e^{iH_{f} t} c_{\sigma} e^{-iH_{f} t} c_{\sigma}^{\dagger} | G \rangle.
\]

This response function is similar to that calculated in the X-ray edge problem[22, 23, 24]: there absorption of an X-ray photon excites an atomic core electron into the conduction band (described by \( c_{\sigma}^{\dagger} \)), leaving behind a core hole which constitutes a scattering potential with respect to \( H_{e} \) (described by \( H_{SC} \)). The calculation of Eq. (S25) is standard (e.g. [24]) and yields (we show only the leading power law)

\[
G_{cc}^{\sigma}(t) \sim t^{-[|\delta_{\sigma} - \pi|^{2} + 2 / \pi^{2}}.
\]

where \( \delta_{\sigma} = \delta(0) \) denotes the phase shifts at the Fermi energy. This power-law has an instructive interpretation, due to Hopfield[21], according to Anderson’s orthogonality catastrophe[13], two Fermi seas subject to different local scattering potentials that cause their modes to differ in phase by \( \delta_{\sigma}(\varepsilon_{k\sigma}) \), have a ground state overlap

\[
\langle i | G | G \rangle \sim L^{-\sum_{\sigma} \delta_{\sigma}^{2}(0) / \pi^{2}} \text{ which vanishes in the limit of system size } L \to \infty.
\]

In analogy, Eq. (S26) can be viewed as the overlap \( \langle i | G | G \rangle \) for systems of size \( L \sim t \) \( \times \text{ the effect of } c_{\sigma}^{\dagger} \), which puts an extra spin-\( \sigma \) electron at the scattering site at \( t = 0 \), is analogous to having an additional infinitely strong scatterer of \( \sigma \)-electrons in the initial, but not the final state, implying an extra shift \(-\pi\) for the phase difference of the \( \delta_{\sigma} \) modes.

The phase shifts at the Fermi energy, needed in Eq. (S26), are given by \( \delta_{\sigma} = \pi \Delta_{\sigma e} \), according to the Friedel sum rule[24, 31], valid for \( T = 0 \) and for arbitrary values of \( B, n_{e}^{L} \) and \( n_{e}^{R} \). Collecting results, we find from Eqs. (S24) and (S26) that

\[
A_{\sigma}^{SC}(\nu) \sim T_{K}^{-1}(\nu / T_{K})^{-\eta_{\sigma}},
\]

with the infrared singularity exponent \( \eta_{\sigma} \) given by Eq. (8),

\[
\eta_{\sigma} = 2 \Delta_{\sigma e} - \sum_{\sigma'} (\Delta_{\sigma e})^{2}.
\]

The dimensionful prefactor in Eq. (S27) was adjusted to ensure that Eqs. (7) and (9) match, up to numerical prefactors, at the crossover scale \( \nu = T_{K} \), implying a prefactor \( (\rho / \Gamma) T_{K}^{-\eta_{\sigma}} \) in Eq. (S29). We have checked Eq. (8) numerically for a range of parameter combinations, see Fig. 5b and Fig. 63 finding it to hold to within 1 %.
In particular, this implies the simple relation
\[ m = \pm \text{sgn}(g_e B) \] (selectable by choice of circular polarization of the incident light), i.e., whether its spin matches that of the lower or upper of the Zeeman-split e-levels, respectively. (Correspondingly, the photoexcited electron will be said to have an “lower”- or “upper-level-spin”, with exponent \( \eta_{\text{lower}} \) or \( \eta_{\text{upper}} \).

This implies a dramatic \( \sigma \)-dependence of the evolution of the lineshape \( A_{\sigma}(\nu) \propto \nu^{-\eta_{\sigma}} \) with increasing \( |B| \) (Fig. 5a). In the main text we focussed explicitly on the case that \( \eta_e = 1 \), for which \( \eta_{\text{lower}}/\eta_{\text{upper}} \) crosses over from \( \frac{1}{2} \) at \( B = 0 \) to \( \pm 1 \) for \( |B| \gg T_K \) (Eq. (9) and Fig. 5b): correspondingly the near-threshold singularity either becomes stronger, tending towards \( \nu^{-1} \); or it becomes weaker, and once \( \eta_{\text{upper}} \) turns negative, changes to an increasingly strong power-law decay, tending toward \( \nu^{+1} \). As mentioned in the main text, the difference between the two cases reflects the fact that in the limit \( |B| \gg T_K \), Anderson orthogonality is absent or maximal for \( \sigma = \) lower or upper, respectively.

In this limit, the dependence of the lineshape on detuning and on \( \sigma \) may also be interpreted in the following, alternative way, in terms of detuned transitions into the e-level: At the absorption edge, the photo-excitation of an upper-level-spin electron can be viewed as being a virtual excitation to the upper Zeeman level (with effective detuning given by \( \sim (\nu - g_e B) \)) followed by a spin-flip of the e-level electron assisted by the creation of a FR electron-hole pair. Since the phase-space for creating these electron-hole pairs scales as \( \nu \), the overall absorption rate for \( \sigma = \) upper scales as \( \nu/\nu^{2} = 1/\nu \). This leads to an absorption rate for \( \sigma = \) lower that scales as \( \nu/\nu^{2} = 1/\nu \). This explains the difference in power laws, \( \eta_{\text{lower}} - \eta_{\text{upper}} = 2 \).

The same picture works for larger photon energies above the edge, except that for the upper-level-spin electron the energy deficit in the virtual state gets smaller. Once the energy deficit reaches zero, a resonant peak in the absorption results. Its width is determined by the Korringa relaxation rate for the upper e-level in the dot. In the scaling regime, the Korringa relaxation rate at large Zeeman energy \( (|g_e B| \gg T_K) \) is of the order of \( g_e B/\ln^{2}(g_e B/T_K) \). Thus, the smaller the exchange interaction with the band (i.e. the smaller \( T_K \)), the sharper the upper-level-spin absorption peak, and the smaller the weight outside the peak, near the absorption edge.

**APPENDIX 7: MAGNETIC FIELD DEPENDENCE OF THE ABSORPTION LINESHAPE**

In the main text, we pointed out that as \( |B| \) increases past \( T_K \), the evolution of the infrared exponents \( \eta_{\sigma} \) with \( B \) depends on whether the spin \( \sigma \) of the photoexcited electron matches that of the lower or upper of the Zeeman-split e-levels, which we distinguished by writing \( \sigma = \) lower or upper, respectively. Here we discuss this \( \sigma \)-dependence in some more detail.

It is instructive to express \( \eta_{\sigma} \) of Eq. (8) in terms of the final occupation \( \bar{n}_{\sigma} \) and magnetic moment \( m_{\sigma} \) of the e-level, writing \( \bar{n}_{\sigma} = \frac{1}{2} \bar{n} + \sigma m_{\sigma} \):

\[ \eta_{\sigma} = n_{\sigma} + 2m_{\sigma}^{2} - 2(m_{\sigma})^{2} \quad \eta_{\sigma} = \bar{n}_{\sigma}(1 - \frac{1}{2} \bar{n}_{\sigma}) \quad \text{(S29)} \]

In particular, this implies the simple relation \( m_{\sigma} = \frac{1}{2}(\eta_{+} - \eta_{-}) \). Moreover, at \( \bar{n}_{e} = 1 \), \( m_{e} \) is a universal function of \( g_e B/T_K \); hence, the same is true for the infrared singularity exponents \( \eta_{\sigma} \). (At very large fields, however, a bulk Zeeman field, neglected above, will spoil universality, see Appendix 8.)

For \( |B| \ll T_K \), the magnetic moment is determined by the linear static susceptibility \( \chi_{0} = 1/4T_K \) via \( m_{\sigma} = -g_e B \chi_{0} \), implying \( \eta_{\sigma} = \eta_{0} - \frac{g_e B}{\chi_{0}} \), to lowest order in \( B/T_K \). As \( |B| \) increases past \( T_K \), and the magnetization tends towards \( m_{\sigma} \approx -\frac{1}{2} \text{sgn}(g_e B) \), the exponents tend to \( \eta_{\sigma} \to -\frac{1}{2}(\bar{n} - 1)^{2} - \frac{\sigma}{2} \text{sgn}(g_e B) \). Thus, they differ by 2 and attain opposite signs (since \( \bar{n}_{e} \in [0, 2] \)), depending on whether the photoexcited electron has spin

\[ \sigma = \pm \text{sgn}(g_e B) \] (selectable by choice of circular polarization of the incident light), i.e., whether its spin matches that of the lower or upper of the Zeeman-split e-levels, respectively. (Correspondingly, the photoexcited electron will be said to have an “lower”- or “upper-level-spin”, with exponent \( \eta_{\text{lower}} \) or \( \eta_{\text{upper}} \).

This implies a dramatic \( \sigma \)-dependence of the evolution of the lineshape \( A_{\sigma}(\nu) \propto \nu^{-\eta_{\sigma}} \) with increasing \( |B| \) (Fig. 5a). In the main text we focussed explicitly on the case that \( \eta_e = 1 \), for which \( \eta_{\text{lower}}/\eta_{\text{upper}} \) crosses over from \( \frac{1}{2} \) at \( B = 0 \) to \( \pm 1 \) for \( |B| \gg T_K \) (Eq. (9) and Fig. 5b): correspondingly the near-threshold singularity either becomes stronger, tending towards \( \nu^{-1} \); or it becomes weaker, and once \( \eta_{\text{upper}} \) turns negative, changes to an increasingly strong power-law decay, tending toward \( \nu^{+1} \). As mentioned in the main text, the difference between the two cases reflects the fact that in the limit \( |B| \gg T_K \), Anderson orthogonality is absent or maximal for \( \sigma = \) lower or upper, respectively.

In this limit, the dependence of the lineshape on detuning and on \( \sigma \) may also be interpreted in the following, alternative way, in terms of detuned transitions into the e-level: At the absorption edge, the photo-excitation of an upper-level-spin electron can be viewed as being a virtual excitation to the upper Zeeman level (with effective detuning given by \( \sim (\nu - g_e B) \)) followed by a spin-flip of the e-level electron assisted by the creation of a FR electron-hole pair. Since the phase-space for creating these electron-hole pairs scales as \( \nu \), the overall absorption rate for \( \sigma = \) upper scales as \( \nu/\nu^{2} = 1/\nu \). This leads to an absorption rate for \( \sigma = \) lower that scales as \( \nu/\nu^{2} = 1/\nu \). This explains the difference in power laws, \( \eta_{\text{lower}} - \eta_{\text{upper}} = 2 \).

The same picture works for larger photon energies above the edge, except that for the upper-level-spin electron the energy deficit in the virtual state gets smaller. Once the energy deficit reaches zero, a resonant peak in the absorption results. Its width is determined by the Korringa relaxation rate for the upper e-level in the dot. In the scaling regime, the Korringa relaxation rate at large Zeeman energy \( (|g_e B| \gg T_K) \) is of the order of \( g_e B/\ln^{2}(g_e B/T_K) \). Thus, the smaller the exchange interaction with the band (i.e. the smaller \( T_K \)), the sharper the upper-level-spin absorption peak, and the smaller the weight outside the peak, near the absorption edge.

**APPENDIX 8: JUSTIFICATION OF NEGLLECTING THE BULK MAGNETIC FIELD**

A magnetic field applied parallel to the Fermi reservoir causes a Zeeman shift not only for the e- and h-levels, but also for the c-electrons in the Fermi reservoir, with \( g \)-factors of comparable magnitude: \( g_{e} \approx -0.6-0.7 \),
A more detailed scaling analysis for $|B| \gg T_K$ is as follows: The effect of a bulk field is to shift the Fermi reservoirs by $-\frac{1}{2} \sigma g_e B$ with respect to the Fermi energy at $\epsilon_F = 0$. We thus used shifted band edges, $D_{\sigma}^{\text{max}}/\min = \pm D - \frac{1}{2} \sigma g_e B$, and employed a $\sigma$-dependent Wilsonian logarithmic energy grid for each of the four energy intervals $[0, D_{\sigma}^{\text{max}}]$ and $[D_{\sigma}^{\text{min}}, 0]$ (with $\sigma = \pm$). For the resulting Wilson chain we calculated the local moment $m_e(B)$ for both $g_e = 0$ and 2, for several different choices of $g_e$, see Fig. S4. As expected, the effect of turning on $g_e$, though noticable for large fields if $g_e = 0$, is very small if $g_e = 2$.

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