

The Kondo exciton: a quantum quench towards strong spin-reservoir correlations

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After a quantum quench, i.e. a sudden change of the Hamiltonian describing a quantum system, its subsequent dynamics is governed by energy scales that become ever lower with increasing time. Time- or frequency-resolved probes of this evolution thus offer insight into how correlations emerge with decreasing energy. Here we study a quantum quench for a semiconductor quantum dot coupled to a Fermionic reservoir, induced by the sudden creation of an exciton via optical absorption. The subsequent emergence of correlations between spin degrees of freedom of dot and reservoir, culminating in the Kondo effect, can be read off from the absorption lineshape and understood in terms of the three fixed points of the single-impurity Anderson model. At low temperatures the lineshape is dominated by a power-law singularity, with an exponent that depends on gate voltage and, in a universal, asymmetric

fashion, on magnetic field, indicative of a tunable Anderson orthogonality catastrophe.

When a quantum dot (QD) is tunnel-coupled to a Fermionic reservoir (FR) and tuned such that its topmost occupied level harbors a single electron, it exhibits at low temperatures one of the most spectacular phenomena of many-body physics – the Kondo effect^{1,2}. In transport studies³⁻⁵, the latter manifests itself via a zero-bias anomaly in the conductance, which reflects the presence of strong correlations between the spin degrees of freedom of the QD and FR, leading to a fully screened spin singlet at $T = 0$. A quantum quench into a parameter regime conducive to such Kondo correlations would allow detailed insight into their emergence as a function of increasing time or decreasing energy. Whereas the transient behavior right after the quench depends on high-energy excitations, the asymptotic long-time evolution is determined by low-lying excitations close to the final ground state and would hence bear distinct signatures of Kondo correlations. Corresponding predictions⁶⁻¹⁰ were made in the context of transport experiments.

Optical transitions in quantum dots¹¹⁻¹³ offer an alternative arena for studying Kondo correlations. Since the creation or annihilation of an exciton via photon absorption or emission is accompanied by a sudden change in the local charge configuration and a sudden switch-on or switch-off of a strong electron-hole attraction¹²⁻¹⁴, an optical transition in effect implements a quantum quench on the QD Hamiltonian. The quench will cause an abrupt change in the nature of any tunnel-induced QD-FR correlations, leaving tell-tale signatures in the absorption and emission lineshapes. For example, at low temperatures and small detunings relative to the threshold, the lineshape has been predicted to show a gate-tunable power-law singularity¹⁴. Though optical sig-

natures of Kondo correlations have not been experimentally observed to date, prospects improved recently due to two key experimental advances: voltage control of the exciton-FR spin dynamics was demonstrated¹⁵, and this was observed to produce non-Lorentzian optical line shapes¹⁶. Although the QD-FR tunnel coupling in these papers was not strong enough to reach the Kondo regime, the latter is certainly within reach of current technology.

In the present paper, we propose a realistic scenario for an optically-induced quantum quench into a regime of strong Kondo correlations. We spell out their experimentally observable signatures in detail by making comprehensive predictions for the absorption lineshape over the entire frequency regime, as function of temperature, gate voltage and magnetic field. In particular, we show in detail that the emergence of Kondo correlations proceeds via three distinct regimes, which are clearly mapped out in the absorption lineshape with decreasing detuning (or, in the time domain, increasing time): (i) For large detuning (short times), charge fluctuations dominate; (ii) for intermediate detuning (intermediate times), spin fluctuations dominate; (iii) for small detuning (long times), the local spin is fully screened and X-ray edge type physics dominates. These three regimes are associated with the three fixed points of the single-impurity Anderson model (AM)¹⁷. The fact that they all leave their trace in the lineshape illustrates, in paradigmatic fashion, the power of a quantum quench to probe spectral information on all energy scales.

The evolution towards strong correlations between a system (the QD) and its environment (the FR) is particularly intriguing and challenging when viewed from the perspective of quantum optics. The FR is a non-standard reservoir that can not be treated using Born and Markov ap-

proximations, because these approximations cannot capture the ensuing many-body correlations between system and environment. Indeed, such approximations would fail completely for regimes (ii) and (iii), where the dynamics is non-Markovian since the spin-excitations of the FR are long-lived. In our analysis, we show how the techniques of quantum optics for calculating optical lineshapes can be extended to correctly capture such correlations, by using the simple but powerful analytical method of fixed-point perturbation theory (FPPT): it exploits the fact that the effects of strong correlations can be encoded in simple effective Hamiltonians, describing the model's renormalization group fixed points. Remarkably, the results of such an analysis are in full quantitative agreement with our numerical results, obtained by the highly reliable, though much more involved, numerical renormalization group (NRG).

Model for a tunable, optically active quantum impurity. We consider a QD, tunnel-coupled to a FR, whose charge state is controllable via an external gate voltage V_g applied between a top Schottky gate and the FR (see Fig. 1 and Supplementary Discussion 1 for details). In a gate voltage regime for which the QD is initially uncharged, a circularly polarized light beam at a suitably chosen frequency ω_L propagating along the z -axis of the heterostructure will create a so-called neutral exciton¹⁸ (X^0), a bound electron-hole pair with well-defined spins σ and $\bar{\sigma} = -\sigma$ ($\in \{+, -\}$) in the localized s -orbitals of the QD's conduction- and valence bands (to be called e- and h-levels, with creation operators $e_\sigma^\dagger$ and $h_{\bar{\sigma}}^\dagger$, respectively). The QD-light interaction is described by $H_L \propto (e_\sigma^\dagger h_{\bar{\sigma}}^\dagger e^{-i\omega_L t} + \text{h.c.})$. We model the system before/after absorption by the initial/final

Hamiltonian $H^{i/f} = H_{\text{QD}}^{i/f} + H_c + H_t$, where

$$H_{\text{QD}}^a = \sum_{\sigma} \varepsilon_{e\sigma}^a n_{e\sigma} + U n_{e\uparrow} n_{e\downarrow} + \delta_{af} \varepsilon_{h\bar{\sigma}} \quad (a = i, f) \quad (1)$$

describes the QD, with Coulomb cost U for double occupancy of the e-level, $n_{e\sigma} = e_{\sigma}^{\dagger} e_{\sigma}$, and hole energy $\varepsilon_{h\bar{\sigma}} (> 0$, on the order of the band gap). The e-level's initial and final energies before and after absorption, $\varepsilon_{e\sigma}^a$ ($a = i, f$), differ by the Coulomb attraction $U_{\text{eh}} (> 0)$ between the newly created electron-hole pair, which pulls the final e-level downward, $\varepsilon_{e\sigma}^f = \varepsilon_{e\sigma} - \delta_{af} U_{\text{eh}}$ (Fig. 1b). This stabilizes the excited electron against decay into the FR, provided that $\varepsilon_{e\sigma}^f$ lies below the FR's Fermi energy $\varepsilon_F = 0$. Since $H^f \neq H^i$, absorption implements a *quantum quench*, which, as elaborated below, can be tuned by electric and magnetic fields. Furthermore, $H_c = \sum_{k\sigma} \varepsilon_{k\sigma} c_{k\sigma}^{\dagger} c_{k\sigma}$ represents a noninteracting conduction band (the FR) with half-width $D = 1/(2\rho)$ and constant density of states $\rho(\varepsilon_k) = \rho\theta(D - |\varepsilon_k|)$ per spin, while $H_t = \sqrt{\Gamma/\pi\rho} \sum_{\sigma} (e_{\sigma}^{\dagger} c_{\sigma} + \text{h.c.})$, with $c_{\sigma} = \sum_k c_{k\sigma}$, describes its tunnel-coupling to the e-level, giving it a width Γ . A magnetic field B along the growth-direction of the heterostructure (Faraday configuration) causes a Zeeman splitting, $\varepsilon_{e\sigma} = \varepsilon_e + \frac{1}{2}\sigma g_e B$, $\varepsilon_{h\sigma} = \varepsilon_h + \frac{3}{2}\sigma g_h B$ (see Supplementary Discussion 1; the Zeeman splitting of FR states can be neglected for our purposes, see Supplementary Discussion 7.) We set $\mu_B = \hbar = k_B = 1$, give energies in units of $D = 1$ throughout, and assume $T, B \ll \Gamma \ll U, U_{\text{eh}} \ll D \ll \varepsilon_{h\bar{\sigma}}$. Supplementary Discussion 1 gives a set of realistic parameters, discusses the range of validity of our model, and explains why it need not explicitly include the effects of electron-hole recombination (very small rate) and intra-dot electron-hole exchange. The latter is anisotropic, which would spoil Kondo screening; however, its effects can be fully compensated by applying a magnetic field fine-tuned to a value, say $B_{\text{eh}}^{\bar{\sigma}}$, that restores degeneracy of the

e-level's two spin configurations (see Fig. S2). In the text below, B is understood to be measured relative to $B_{\text{eh}}^{\bar{\sigma}}$.

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, $\bar{n}_e^i \simeq 0$ and $\bar{n}_e^f \simeq 1$. (Here $\bar{n}_e^a = \sum_{\sigma} \bar{n}_{e\sigma}^a$, and $\bar{n}_{e\sigma}^a = \langle n_{e\sigma} \rangle_a$ is the thermal average of $n_{e\sigma}$ with respect to H^a .) This requires $\varepsilon_{e\sigma}^i \gg \Gamma$, and $-U + \Gamma \lesssim \varepsilon_{e\sigma}^f \lesssim -\Gamma$. The initial ground state, needed below, will thus be approximated by the free Fermi sea, $|G\rangle_i \simeq \prod_{\varepsilon_{k\sigma} < \varepsilon_F} c_{k\sigma}^\dagger |\text{Vac}\rangle$, neglecting terms of order $\Gamma/\varepsilon_{e\sigma}^i$. 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 $\varepsilon_{e\sigma}^f = -U/2$, for which $\bar{n}_e^f = 1$ 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 exciton at time $t = 0$, it is instructive to calculate the subsequent time evolution (defined in Methods) of the average charge $\tilde{n}_e(t) = (\tilde{n}_{e+} + \tilde{n}_{e-})(t)$ and spin polarization $\tilde{m}_e(t) = \frac{1}{2}(\tilde{n}_{e+} - \tilde{n}_{e-})(t)$ of the e-level. (The hole has no dynamics; its only role is to pull the e-level down and implement a quench.)

Fig. 2 shows a typical result for $T = 0$ and $H^f=\text{SEAM}$, obtained using time-dependent NRG^{9,10}.

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 $t \simeq 1/|\varepsilon_{e\sigma}^f|$ ($= 2/U$ for $H^f=\text{SEAM}$) associated with virtual transitions of electrons between e-level and FR. Whereas the charge equilibrates (towards 1) shortly thereafter, (ii) the spin decays (towards $\simeq 0$) much more slowly, on the scale $t \simeq 1/T_K$, where $T_K = \sqrt{\Gamma U/2} e^{-\pi|\varepsilon_e^f(\varepsilon_e^f+U)|/(2U\Gamma)}$ is the Kondo temperature¹⁹

associated with H^f . The decay is due to spin-flip processes, mediated by electrons of opposite spin hopping between e-level and FR, leading to non-Markovian dynamics because the bath remembers its spin state between two e-level spin-flips. As a result a FR screening cloud builds up over time, which ultimately screens the localized spin into a singlet.

The time-evolution depicted in Fig. 2 could in principle be observed by a π -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 ω_L exceeds a threshold frequency $\omega_{th} = E_G^f - E_G^i$, which is on the order of $\varepsilon_{e\sigma}^f + \varepsilon_{h\bar{\sigma}}$ (minus corrections due to tunneling and correlations). By Fermi's golden rule the absorption lineshape at temperature T and detuning $\nu = \omega_L - \omega_{th}$ is proportional to the spectral function $A_\sigma(\nu) = \mathcal{A}_\nu[e_\sigma^\dagger; H^f]$, where we use the general notation

$$\mathcal{A}_\nu[\hat{X}^\dagger; H^f] = 2\pi \sum_{mm'} \rho_m^i \left| {}_f\langle m' | \hat{X}^\dagger | m \rangle_i \right|^2 \delta(\omega_L - E_{m'}^f + E_m^i) \quad (2)$$

for the spectral function of the observable $\hat{X}^\dagger$ with respect to the final and initial Hamiltonians H^f and H^i (the dependence on the latter is implied but will not be displayed.) Here $|m\rangle_a$ and E_m^a are the exact eigenstates and -energies of H^a and $\rho_m^i = e^{-E_m^i/T}/Z^i$ the initial Boltzmann weights. At $T = 0$, Eq. (2) reduces to

$$\mathcal{A}_\nu[\hat{X}^\dagger; H^f] = -2 \text{Im} {}_i\langle G | \hat{X} \frac{1}{\nu_+ - \bar{H}^f} \hat{X}^\dagger | G \rangle_i, \quad (3)$$

with $\nu_+ = \nu + i0_+$ and $\bar{H}^f = H^f - E_G^f$. Note that Eq. (3) equals $-2 \text{Im} \int_0^\infty dt e^{it\nu_+} \langle \Psi(0) | \Psi(t) \rangle$, with $|\Psi(0)\rangle = \hat{X}^\dagger |G\rangle_i$ and $|\Psi(t)\rangle = e^{-i\bar{H}^f t} |\Psi(0)\rangle$. Thus the absorption lineshape $A_\sigma(\nu)$, for which $\hat{X}^\dagger = e_\sigma^\dagger$, directly probes the post-quench dynamics, with initial state $e_\sigma^\dagger |G\rangle_i$, of a photo-generated e-electron coupled to a FR.

We used NRG to calculate $A_\sigma(\nu)$ from Eq. (2), generalizing the approach of Ref. 14 to $T \neq 0$ by following Ref. 20 (see Methods and Supplementary Discussion 2 for details). For clarity, we focus first on H^f =SEAM with $B = 0$. Fig. 3a shows a typical result: As temperature is gradually reduced, an initially rather symmetric lineshape becomes highly asymmetric, dramatically increasing in peak-height as $T \rightarrow 0$. At $T = 0$, the lineshape displays a threshold, vanishing for $\nu < 0$ and *diverging* as ν 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. 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:

$$\text{(FO)} \quad |\varepsilon_{e\sigma}^f| \lesssim \nu \lesssim D \quad : \quad A_\sigma^{\text{FO}}(\nu) = \frac{4\Gamma}{\nu^2} \theta(\nu - |\varepsilon_{e\sigma}^f|) ; \quad (4a)$$

$$\text{(LM)} \quad T_K \lesssim \nu \lesssim |\varepsilon_{e\sigma}^f| \quad : \quad A_\sigma^{\text{LM}}(\nu) = \frac{3\pi}{4\nu} \ln^{-2}(\nu/T_K); \quad (4b)$$

$$\text{(SC)} \quad T \lesssim \nu \lesssim T_K \quad : \quad A_\sigma^{\text{SC}}(\nu) \propto T_K^{-1} (\nu/T_K)^{-\eta_\sigma} . \quad (4c)$$

The remarkable series of crossovers found above are symptomatic of three different regimes of charge and spin dynamics that cannot be described by standard quantum optical techniques. Below we harness Wilson’s concept^{17,21} of associating different dynamical regimes with different renormalization group (RG) fixed points of the model to propose a simple analytical approach for

calculating spectral functions, using *fixed point perturbation theory* (FPPT). We show that this scheme correctly captures the behavior of the spectral function in all three dynamical regimes. To this end we note that the absorption spectrum at large, intermediate or small detuning probes excitations at successively smaller energy scales (see Fig. 1c), corresponding to ever longer time scales after absorption, for which H^f can be represented by expansions $H_r^* + H_r'$ around the three well-known fixed points¹⁷ of the AM: the *free orbital*, *local moment* and *strong-coupling* fixed points ($r = \text{FO, LM, SC}$), characterized by charge fluctuations, spin fluctuations and spin screening, respectively. By focussing, at each energy scale, on the relevant degrees of freedom (described by H_r^*) and their mutual interactions (described by H_r'), FPPT captures the complexity of the system's many-body correlations in a simple and efficient manner.

Large and intermediate detuning – perturbative regime. For large detuning, probing the time interval $t \lesssim 1/|\varepsilon_{e\sigma}^f|$ immediately after absorption, the e-level appears as a free, filled orbital perturbed by charge fluctuations, described by¹⁷ the fixed point Hamiltonian $H_{\text{FO}}^* = H_c + H_{\text{QD}}^f$ and the relevant perturbation $H_{\text{FO}}' = H_t$. Intermediate detuning probes the times $1/|\varepsilon_{e\sigma}^f| \lesssim t \lesssim 1/T_K$ for which real charge fluctuations have frozen out, resulting in a stable local moment; however, virtual charge fluctuations still cause the local moment to undergo spin fluctuations, which are not yet screened. This is described by¹⁷ $H_{\text{LM}}^* = H_c + \text{const.}$ and the RG-relevant perturbation $H_{\text{LM}}' = \frac{J(\nu)}{\rho} \vec{s}_e \cdot \vec{s}_c$ (a potential scattering term in H_{LM}' , being RG-irrelevant, will be neglected in the discussion of the intermediate-detuning regime). Here $\vec{s}_j = \frac{1}{2} \sum_{\sigma\sigma'} j_\sigma^\dagger \vec{\tau}_{\sigma\sigma'} j_{\sigma'}$ (for $j = e, c$) are spin-operators for the e-level and conduction band, respectively ($\vec{\tau}$ are Pauli matrices), and

$J(\nu) = \ln^{-1}(\nu/T_K)$ is an effective, scale-dependent dimensionless exchange constant. Constant contributions to H_r^* will not be specified, since they affect only ω_{th} , whose precise value is not of present interest. It suffices to note that for both $r = \text{FO}$ and LM , $e_\sigma^\dagger|G\rangle_i$ is an eigenstate of H_r^* with eigenvalue E_G^f (within the accuracy of H_r^*).

For $r = \text{FO}$ and LM , $A_\sigma(\nu)$ can be calculated using perturbation theory in H_r' . For $T = 0$, we set $\bar{H}^f \rightarrow \bar{H}_r^* + H_r'$ in Eq. (3) (with $\hat{X}^\dagger = e_\sigma^\dagger$) and make a resolvent expansion of $(\nu_+ - \bar{H}_r^* - H_r')^{-1}$ in powers of H_r' . To lowest non-vanishing order, Eq. (3) then reduces to (excluding a $\delta(\nu)$ term not relevant for $\nu \gtrsim T_K$):

$$A_\sigma^r(\nu) \simeq \nu^{-2} \mathcal{A}_\nu[H_r' e_\sigma^\dagger; H_r^*] . \quad (5)$$

This structure reveals the relevant physics with striking clarity. Large detuning ($r = \text{FO}$) is described by the spectral function of the operator $H_t e_\sigma^\dagger$; the absorption process can thus 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. In contrast, intermediate detuning ($r = \text{LM}$) is described by the spectral function of $\vec{s}_c \cdot \vec{s}_e e_\sigma^\dagger$, i.e. it probes spin fluctuations, as observed in the dynamics (Fig. 2). Evaluation of these spectral functions is elementary since H_{FO}^* and H_{LM}^* involve only free fermions. For $B = 0$ and $|\varepsilon_{e\sigma}^f| = \frac{1}{2}U$, we readily recover Eqs. (4a) and (4b), which quantitatively agree with the NRG results of Fig. 3b.

Depending on the detailed choice of parameters, the crossover from LM to FO shows up in our numerical results as a small shoulder or side peak in $A_\sigma(\nu)$ at $|\varepsilon_{e\sigma}^f|$ (easily seen from the fact that $A_\sigma^{\text{LM}}(|\varepsilon_{e\sigma}^f|) \neq A_\sigma^{\text{FO}}(|\varepsilon_{e\sigma}^f|)$). This crossover reflects the following facts: Transitions from the

h-level into unfilled states of the FR are possible only for $\nu > |\varepsilon_{e\sigma}^f|$ (hence the step function in Eq. (4a)); they are mediated by H'_{FO} and use the e-level (Lorentzian-broadened by charge fluctuations, hence the ν^{-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'_{LM} -induced creation of electron-hole pairs in the FR, causing $A_{\sigma}^{\text{LM}}(\nu)$ of Eq. (4b) to have an additional phase space factor $\propto \nu$ relative to $A_{\sigma}^{\text{FO}}(\nu)$ of Eq. (4a).

The FPPT strategy for calculating FO and LM lineshapes can readily be generalized to finite temperatures (the details follow, in part, Ref. 22). For example, for $T_{\text{K}} < |\nu| < |\varepsilon_{e\sigma}^f|$ and $T \gg T_{\text{K}}$ we obtain

$$A_{\sigma}^{\text{LM}}(\nu) = \frac{3\pi}{4} \frac{\nu/T}{1 - e^{-\nu/T}} \frac{\gamma_{\text{Kor}}(\nu, T)/\pi}{\nu^2 + \gamma_{\text{Kor}}^2(\nu, T)}. \quad (6)$$

Here $\gamma_{\text{Kor}}(\nu, T) = \pi T / \ln^2[\max(|\nu|, T)/T_{\text{K}}]$ is the scale-dependent Korringa relaxation rate^{2,22}. It is *smaller* than T by a large logarithmic factor, implying that the absorption peak is considerably narrower and higher than for the usual case of thermal broadening. For $\nu > T$, Eq. (6) reduces to Eq. (4b). A comparison of Eq. (6) to NRG results is given in Fig. 3c and its caption.

For clarity, the above discussion was confined to $H^f = \text{SEAM}$. However, it can be generalized straightforwardly to the non-symmetric case with $\varepsilon_{e\sigma}^f \neq -\frac{1}{2}U$, as long as H^f remains in the LM-regime, with $\bar{n}_e^f \simeq 1$ (see Supplementary Discussion 4). The lineshape then depends on $\varepsilon_{e\sigma}^f$ and U only through their influence on T_{K} , and hence $A_{\sigma}^{\text{LM}}(\nu)$ is a *universal* function of ν and T_{K} . This is illustrated in Fig. 3d for five lineshapes, shown in the lower left panel, corresponding to different choices of $\varepsilon_{e\sigma}^f$ and hence different T_{K} -values (as indicated in inset). When these lineshapes are

rescaled as $A_\sigma(\nu)/A_\sigma(T_K)$ vs. ν/T_K (main panel), the ones with $\bar{n}_{e\sigma}^f \simeq 1$ (green, red, blue) collapse onto a universal scaling curve within the LM regime $T_K \lesssim \nu \lesssim |\varepsilon_{e\sigma}^f|$. An experimental observation of such a scaling collapse would be a smoking gun for the existence of Kondo correlations.

Small detuning and tunable Kondo-edge singularity – strong-coupling regime. As ν is lowered through the bottom of the LM regime, $J(\nu)$ increases through unity into the strong coupling regime, and $A_\sigma(\nu)$ monotonically crosses over to the SC regime, first studied for the present model by Helmes *et al.*¹⁴. In this regime the FR is strongly altered by the interactions with the QD (implying the complete inapplicability of a Born-Markov treatment). As mentioned above in the discussion of Fig. 2, on a timescale $1/T_K$ a screening cloud builds up that tends to screen the local moment into a spin singlet, (see the decay of $\tilde{m}_e(t \rightarrow \infty) \rightarrow 0$ in Fig. 2). The screened spin singlet acts as a source of strong potential scattering for other FR electrons. Its emergence will appear to be sudden when viewed at very long times ($t \gg 1/T_K$), corresponding to small detuning, in which case analogies can be drawn to the X-ray edge problem^{23,24}. The latter describes the absorption of an incident X-ray via the sudden transfer of an electron from an atomic core level into the conduction band. The remaining core hole is a source of potential scattering for other FR electrons, causing their phases to shift. This leads to the initial and final FR ground states to be orthogonal to each other (Anderson’s orthogonality catastrophe²⁵), and a power-law singularity to arise in the X-ray absorption lineshape. Studying the Kondo exciton absorption line shape for small detuning and $B = 0$, Helmes *et al.* found a similar singularity, of the form (4c); moreover, its exponent η_σ was shown to be *tunable* via the gate voltage in a way described by “Hopfield’s rule of thumb” (put

forth in Ref. 26 for the X-ray problem). We show now that η_σ also depends on magnetic field, in a remarkably B -asymmetric fashion, reflecting the fact that once the incident light's polarization is fixed, opposite field directions become inequivalent.

Generalizing Hopfield's rule (see Methods) to the case of $B \neq 0$, for which spin symmetry is broken, we find

$$\eta_\sigma = 2\Delta n_{e\sigma} - \sum_{\sigma'} (\Delta n_{e\sigma'})^2, \quad (7)$$

where $\Delta n_{e\sigma} = \bar{n}_{e\sigma}^f - \bar{n}_{e\sigma}^i$ is the change in the spin-resolved e-level's average occupation. Supplementary Discussion 5 offers a detailed derivation of Eq. (7) that puts the heuristic arguments of Helmes *et al.* on a rigorous footing using FPPT in the time domain. Eq. (7) 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 $t \rightarrow \infty$) and the initial state $|0^+\rangle$ (at $t = 0^+$) just after photo-excitation of a spin- σ electron. The "1" in η_σ represents a ν^{-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 ν) to carry off the excess energy ν , resulting in a lineshape scaling as $\nu/\nu^2 = \nu^{-1}$. The $\sum_{\sigma'} (\Delta n'_{e\sigma'})^2$ contribution to η_σ reflects Anderson orthogonality²⁵: 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, implying²⁵ that their overlap scales with effective system size $L \sim \nu^{-1}$ as $\langle \infty | 0^+ \rangle \sim L^{-\sum_{\sigma'} (\Delta n_{e\sigma'})^2}$.

According to Eq. (7), η_σ can be tuned experimentally not only via gate voltage but also via

magnetic field, since both modify $\varepsilon_{e\sigma}^a$ and hence $\Delta n_{e\sigma'}$ (see upper inset of Fig. 3d, Fig. 4b and Fig. S4). This *tunability* can be exploited to study universal aspects of Anderson orthogonality physics that had hitherto been inaccessible. In particular, if the system is tuned such that $\bar{n}_e^i = 0$ and $\bar{n}_e^f = 1$, Eq. (7) can be expressed as

$$\eta_\sigma = \frac{1}{2} + 2m_e^f \sigma - 2(m_e^f)^2, \quad (8)$$

where the final magnetization $m_e^f = \frac{1}{2}(\bar{n}_{e+}^f - \bar{n}_{e-}^f)$ is a universal function of $g_e B/T_K$. (At very large fields, however, a bulk Zeeman field, neglected above, will spoil universality, see Supplementary Discussion 7.) The exponents η_σ then are *universal functions of $g_e B/T_K$* , with simple limits for small and large fields (see Fig. 4b):

$$\eta_{\text{lower/upper}} \rightarrow \begin{cases} \frac{1}{2} & (|g_e B| \ll T_K), \\ \pm 1 & (|g_e B| \gg T_K). \end{cases} \quad (9)$$

The subscript “lower” or “upper” distinguishes whether the spin- σ electron is photo-excited into the “lower” or “upper” of the Zeeman-split pair ($\sigma g_e B < 0$ or > 0 , respectively); reversing the direction of B (at fixed incident photon polarization) will thus interchange “lower” and “upper”. The sign difference between η_{lower} and η_{upper} for $|g_e B| \gg T_K$ arises since the change in local charge becomes fully asymmetric, $\Delta n_{e,\text{lower}} \rightarrow 1$ while $\Delta n_{e,\text{upper}} \rightarrow 0$; as a result, Anderson orthogonality is completely absent ($\Delta n'_{e\sigma'} = 0$) for photo-excitation into the “lower” level, since subsequently the e-level spin need not adjust at all. In contrast, it is maximal ($\Delta n'_{e\sigma'} = 1$) for photo-excitation into the “upper” level, since subsequently the e-level spin has to create a spin-flip electron-hole pair excitation in the FR to reach its long-time value. It follows, remarkably, that a magnetic field tunes the strength of Anderson orthogonality, implying a dramatic *asymmetry* for the evolution

of the lineshape $A_\sigma(\nu) \propto \nu^{-\eta_\sigma}$ with increasing $|B|$ (Fig. 4a): For $A_{\text{lower}}(\nu)$, the near-threshold singularity becomes stronger, tending towards ν^{-1} . In contrast, for $A_{\text{upper}}(\nu)$ the singularity becomes weaker, and once η_{upper} turns negative, changes to an increasingly strong power-law decay, tending toward ν^{+1} ; this is accompanied by the emergence of an absorption peak near $\nu = \sigma g_e B$, associated with a transition into the upper Zeeman-split level, broadened by Korringa relaxation of its spin (see Supplementary Discussion 6). The fact that reversing the direction of B will turn a near-threshold divergence in the lineshape into 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 of the absorption threshold. The *shift* of the absorption threshold frequency $\omega_{\text{th}} = E_G^f - E_G^i$ with magnetic field can be written as $\omega_{\text{th}}(B) - \omega_{\text{th}}(0) = \frac{3}{2}\bar{\sigma}g_h B + \delta\omega_{\text{th}}^e(B)$. The first term reflects the Zeeman energy of the photo-excited hole (which has pseudo-spin 3/2), the second the B -dependence of the ground-state energy of the electron system. The general $T = 0$ relation $g_e m_e^a = \partial E_G^a / \partial B$ implies that the differential threshold shift offers a direct way of experimentally measuring the local moment difference between the final and initial ground states: $\partial(\delta\omega_{\text{th}}^e)/\partial B = g_e[m_e^f(B) - m_e^i(B)]$. Moreover, for $\bar{n}_e^i \simeq 0$ and $\bar{n}_e^f \simeq 1$ the asymptotic behavior of m_e^f for small fields ($m_e^f = -g_e B \chi_0$, where $\chi_0 = 1/(4T_K)$ is the linear static susceptibility) and large fields ($|m_e^f| = \frac{1}{2}$) implies:

$$\delta\omega_{\text{th}}^e = \begin{cases} -(g_e B)^2 / 8T_K & (|B| \ll T_K) , \\ -|g_e B|/2 & (T_K \ll |g_e B| \ll |\varepsilon_e^f|) . \end{cases} \quad (10)$$

The quadratic B -dependence of $\delta\omega_{\text{th}}^{\text{e}}$ for small fields (confirmed numerically in Supplementary Discussion 8) 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.

Conclusion. We have demonstrated that optical absorption in a single quantum dot can be used to implement a *tunable* quantum quench in an experimentally accessible solid-state system. In particular, this allows the Anderson orthogonality catastrophe, a fundamental phenomenon long known in solid state physics, to be studied in a tunable setting. Our work sets the stage for exploring numerous further interesting problems, such as (i) the exploration of the anisotropic exchange interaction between a QD heavy-hole and a FR; (ii) the dynamics following a quantum quench when the FR is an interacting Luttinger liquid; (iii) attempting to exploit strong correlations to realize coherent single-spin rotation or a “Fermionic quantum bus” between two distant QD spins, and (iv) the nonlinear Kondo exciton, where the QD transition is driven by a non-perturbative laser field and an intriguing interplay between Rabi oscillations of the photon-exciton system and Kondo correlations of the exciton-FR system can be expected if the Rabi frequency becomes of the order of the Kondo temperature.

Methods

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 the full density matrix (FDM) numerical renormalization group (NRG)²⁰. 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 ($\{|m\rangle_i\}$ and $\{|m'\rangle_f\}$) as well as eigenenergies ($\{E_m^i\}$ and $\{E_{m'}^f\}$). 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^{9,10}: for each shell k , the contribution towards the initial density matrix ρ^i from that shell (obtained using data from NRG run #1), and the matrix elements $|\langle m' | e_\sigma^\dagger | m \rangle_i|^2$ between shell- k eigenstates from NRG runs #2 and NRG #1 are calculated, and binned according to the corresponding frequency difference $E_{m'}^f - E_m^i$. See Supplementary Discussion 2 for further details, and Table S1 there for a summary of the NRG parameters used for each figure.

Nonequilibrium dynamics via NRG. The expectation value of an observable $\hat{X}$ after absorption is given by $\tilde{X}(t) = \text{Tr}(\hat{\rho}_p^f(t)\hat{X})$ where the time evolution is governed by the final Hamiltonian, $\hat{\rho}_p^f(t) \equiv e^{-iH^f t} \hat{\rho}_p^f e^{iH^f t}$. For reasons discussed in Supplementary Discussion 3 we find it convenient to initialize this evolution with the projected initial density matrix $\hat{\rho}_p^f = e_\sigma^\dagger \left(e^{-H^i/T} / Z^i \right) e_\sigma / [1 - \bar{n}_{e\sigma}]$. The Fourier transform of $\tilde{X}(t)$, $\tilde{X}(\omega) = \int dt e^{i\omega t} \tilde{X}(t)$, can be expressed in Lehmann repre-

sensation:

$$\tilde{\mathcal{X}}(\omega) = \sum_{mm'} {}_f\langle m' | \hat{\rho}_p^f | m \rangle {}_f\langle m | \hat{B} | m' \rangle_f \cdot 2\pi \delta(\omega - E_{m'}^f + E_m^f). \quad (11)$$

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

Absorption in the strong-coupling regime and Fermi-edge physics. The strong-coupling regime features a screened spin singlet that 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^i . This regime can be described by a strong-coupling fixed-point Hamiltonian $H_{\text{SC}}^* + H_{\text{SC}}'$ due to Nozières²⁷ (given in Supplementary Discussion 5). It is formulated purely in terms of these phase-shifted c-electrons and makes no reference to e-level operators at all. Thus, FPPT in the energy domain, as used in the main text, cannot be applied here.

This hurdle can be overcome by working in the time domain: Eq. (2) can be expressed as $\mathcal{A}_\nu[\hat{X}^\dagger; H^f] = -2\text{Im}\mathcal{G}_\nu[\hat{X}, \hat{X}^\dagger]$ via $\mathcal{G}_\nu[\hat{X}, \hat{X}^\dagger] = \int dt e^{it(\omega_{\text{th}} + \nu + i0)} G_t[\hat{X}, \hat{X}^\dagger]$, the Fourier transform (with infinitesimal damping factor) of the correlator

$$G_t[\hat{X}, \hat{X}^\dagger] = -i\theta(t) {}_i\langle e^{iH^i t} \hat{X} e^{-iH^f t} \hat{X}^\dagger \rangle_i. \quad (12)$$

The anomalous time dependence of the latter, involving both H^i and H^f , reflects the fact that the creation of a hole during optical absorption abruptly lowers the e-level. Thus, the desired spectral function $A_\sigma(\nu) = \mathcal{A}_\nu[e_\sigma^\dagger; H^f]$ depends on the nonequilibrium correlator $G_t[e_\sigma, e_\sigma^\dagger]$. The latter can be related to a correlator involving only FR operators, $G_t[c_{k\sigma}, c_{k'\sigma}^\dagger]$, using equations of motion, and subleading terms in the latter can be neglected upon taking the long-time limit. Transforming back to the frequency domain and summing over $\sum_{kk'}$ gives (see Supplementary Discussion 5 for

details):

$$A_{\sigma}^{\text{SC}}(\nu) \sim -\frac{1}{\pi\rho\Gamma}\mathcal{A}_{\nu}[c_{\sigma}^{\dagger}; H_{\text{SC}}^{*}] , \quad (13)$$

where we have set $H^{\text{f}} \rightarrow H_{\text{SC}}^{*}$, in accord with the limit $\nu \rightarrow 0$ (likewise, $H^{\text{i}} \rightarrow H_{\text{c}}$ is implied; the contribution of the perturbation H'_{SC} around the fixed point can be neglected, since H_{SC}^{*} itself gives the dominant contribution). Evidently, absorption in the SC regime can be understood as a transition "directly" into the FR, since the right-hand side of Eq. (13) features a c-electron spectral function. This reflects the strong-correlations inherent in the ground state between the e-level electron and the FR electrons. Indeed, the correlator governing the latter,

$$G_t[c_{\sigma}, c_{\sigma}^{\dagger}] \sim {}_{\text{i}}\langle \text{G} | \text{e}^{iH_{\text{c}}t} c_{\sigma} \text{e}^{-iH_{\text{SC}}^{*}t} c_{\sigma}^{\dagger} | \text{G} \rangle_{\text{i}} , \quad (14)$$

is familiar from the X-ray edge problem^{23,24}. It decays as $\sim t^{-[(\delta_{\sigma}-\pi)^2+\delta_{\sigma}^2]/\pi^2}$ (we show only the leading power law), where δ_{σ} , the phase shifts at the Fermi energy, are found as $\delta_{\sigma} = \pi\Delta n_{\text{e}\sigma}$ from the Friedel sum rule²⁸, valid for $T = 0$ and for arbitrary values of B , $\bar{n}_{\text{e}\sigma}^{\text{f}}$ and $\bar{n}_{\text{e}\sigma}^{\text{i}}$. Collecting results, we find Eq. (4c) and Eq. (7). See Supplementary Discussion 5 for details.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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Figure 1 Schematics of the absorption experiment. The Fermionic reservoir in the back-contact (of e.g. a Schottky-diode structure) couples via charge tunneling to the localized electronic degrees of freedom of the quantum dot (see Supplementary Discussion 1). Starting from **a** an uncharged QD at $t = 0$, optical excitation to a **b** neutral exciton state (X^0) at $t = 0^+$ featuring a single electron in the e-level of the QD (along with a hole in the h-level) which may develop Kondo correlations with the Fermionic reservoir in the long-time limit. (c) Starting from an empty QD state $|G\rangle_i$ (for $T = 0$) the absorption probability of a photon at frequency $\omega_L = \omega_{th} + \nu$, given by $\mathcal{A}_\nu[e_\sigma^\dagger; H^f]$ of Eq. (2), probes the many body spectrum of H^f at excitation energy ν , where $\omega_{th} = E_G^f - E_G^i$ is the absorption threshold. H^f represents the single-impurity Anderson model, which displays three different regimes of dynamical behavior. The final state spectrum $\{|m'\rangle_f, E_{m'}^f\}$ can correspondingly be divided into regimes of “large”, “intermediate” or “small” excitation energies labeled (for reasons discussed in the text) by $r = \text{FO, LM and SC}$, for which H^f can be represented by expansions $H_r^* + H_r'$ around the model’s three fixed points¹⁷. This can be exploited to obtain a fully analytic understanding of the entire range of dynamical behavior.

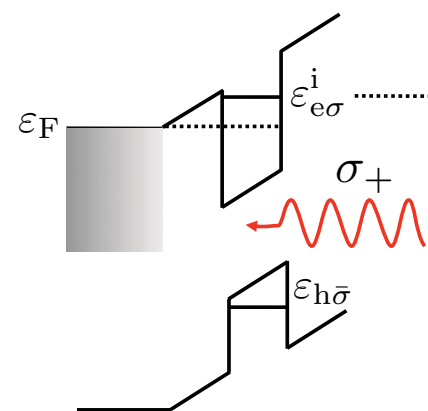
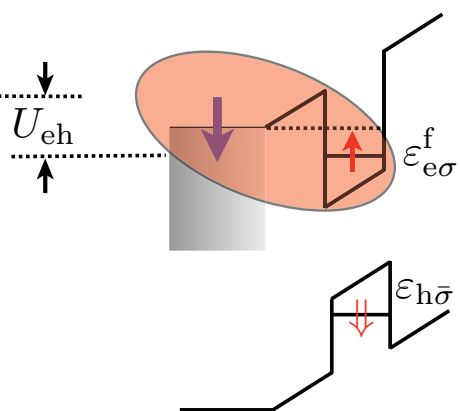
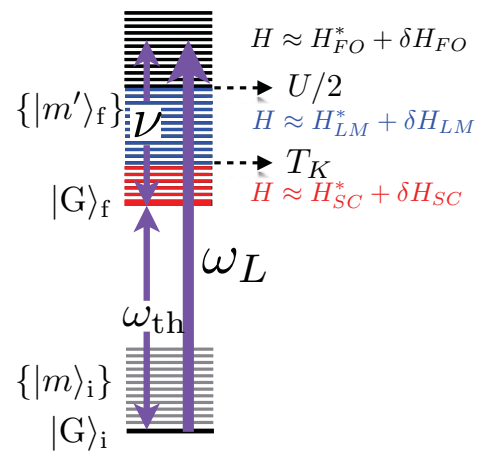
Figure 2 Non-equilibrium time evolution of charge and spin of the photo-excited electron. Plotted is the non-equilibrium time-evolution of charge and spin degrees of freedom of the photo-excited electron after the sudden creation of an $e_+^\dagger h_-^\dagger$ exciton at time $t = 0$. While fluctuations in both e-level’s total charge $\tilde{n}_e$ and spin set in around the time scale $1/|\varepsilon_\sigma^f|$, the equilibration of the spin- σ populations $\tilde{n}_{e\sigma}(t)$ and screening of the local spin $\tilde{m}_e(t)$ sets in on the time scale $1/T_K$. The deviations (by about 3%) of $\tilde{n}_e(\infty)$, $\tilde{n}_{e\sigma}(\infty)$ and $\tilde{m}_e(\infty)$

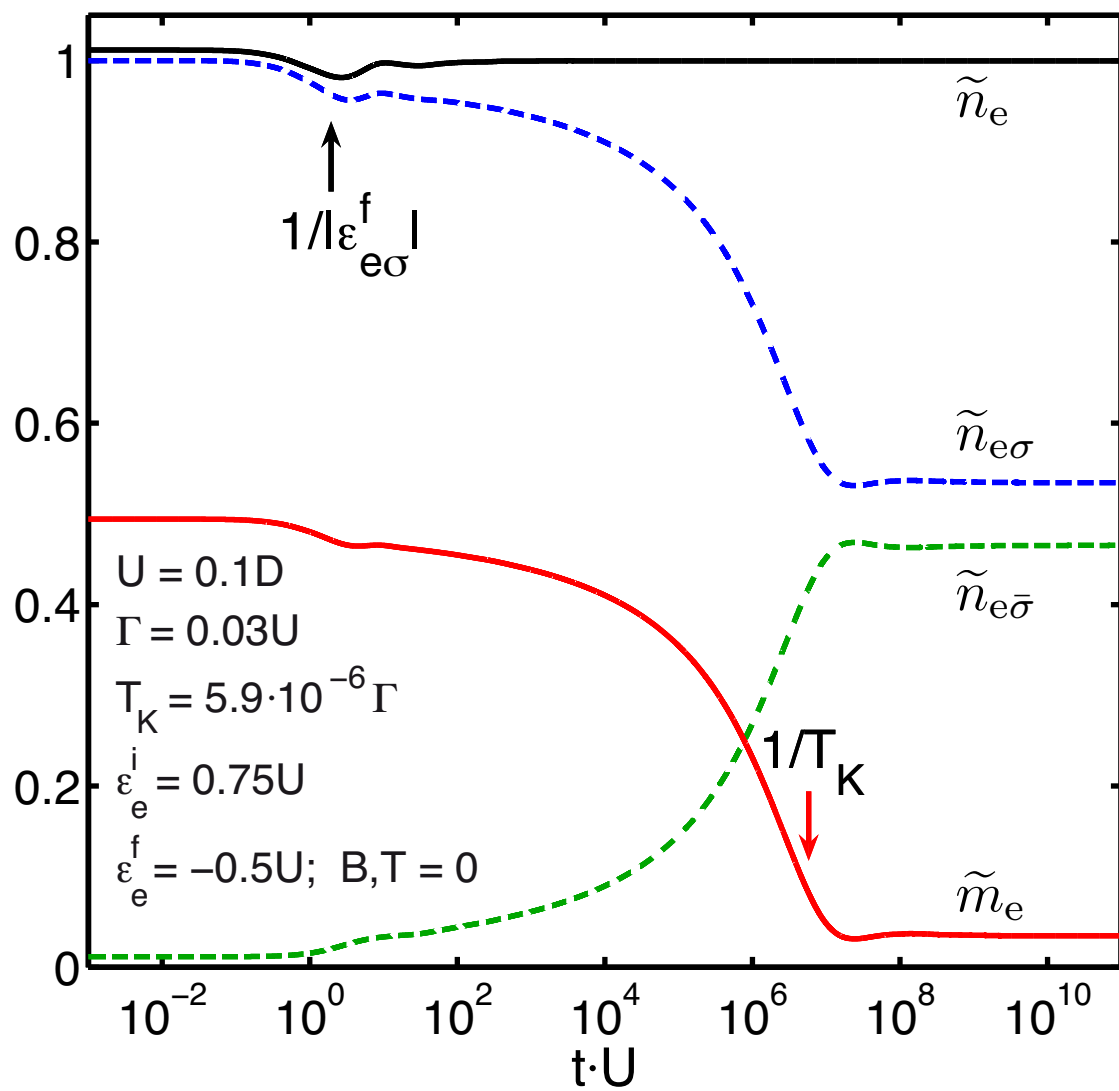
from their expected equilibrium values (1 , $\frac{1}{2}$ and 0 , respectively, for the case $H^f = \text{SEAM}$ depicted here), are known artifacts of time-dependent NRG^{9,10}, presumably due to the NRG discretization scheme, which is inevitably coarse at large energies (see Supplementary Discussion 3). Here and elsewhere, numerical T_K values were determined via $T_K = 1/(4\chi_0)$ from the static linear susceptibility χ_0 .

Figure 3 $B = 0$ absorption lineshape for different temperatures and gate voltages. Solid lines give NRG results for $A_\sigma(\nu)$, calculated (**a,b,c**) for $H^f = \text{SEAM}$ for various temperatures, and (**d**) for different gate voltages at low temperature. Dashed lines show analytical predictions for $\Gamma_i = 0$, for **a,c** from Eq. (6) and for **b** from Eqs. (4). **a**, Semi-log plots for five different temperatures (inset: zoom-in to reveal near-threshold behavior). **b**, Log-log plot (inset: linear plot) for $T \ll T_K$, showing three distinct functional forms for high, intermediate and small detuning, labeled FO, LM and SC, respectively, according to the corresponding fixed points of the Anderson model. Arrows and light yellow lines indicate the crossover scales T , T_K and $|\varepsilon_{e\sigma}^f|$. **c**, Log-log plot (inset: log-linear plot) for $T \gg T_K$ ($\gamma_{\text{Kor}} = \gamma_{\text{Kor}}(0, T)$). For $\nu \gtrsim 0.5T$, the good agreement in **b,c** between analytics (red dashed lines) and NRG (thick blue line for $\Gamma_i \neq 0$; thin blue line for $\Gamma_i = 0$, in **b** only) over many orders of magnitude illustrates the quantitative accuracy of FPPT. For $\nu \lesssim 0.5T$ (**c**, inset), NRG is not quantitatively accurate (the spurious dip at $\nu = 0$ is explained in Supplementary Discussion 2, see also Fig. S3); neither is Eq. (6), unless $\ln(T/T_K) \gg 1$ (which is not the case in **c**), because we specified $\gamma_{\text{Kor}}(\nu, T)$ only with logarithmic accuracy. **d**, Universality in the LM-regime. Upper inset: $\bar{n}_e^i$, $\bar{n}_e^f$, Δn_e and T_K as functions of

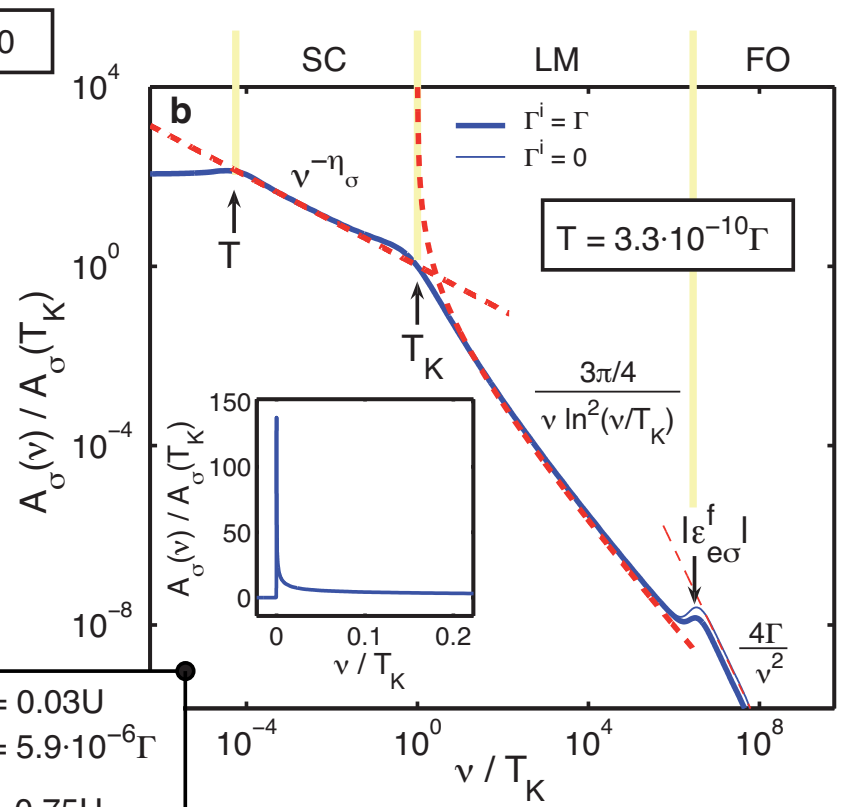
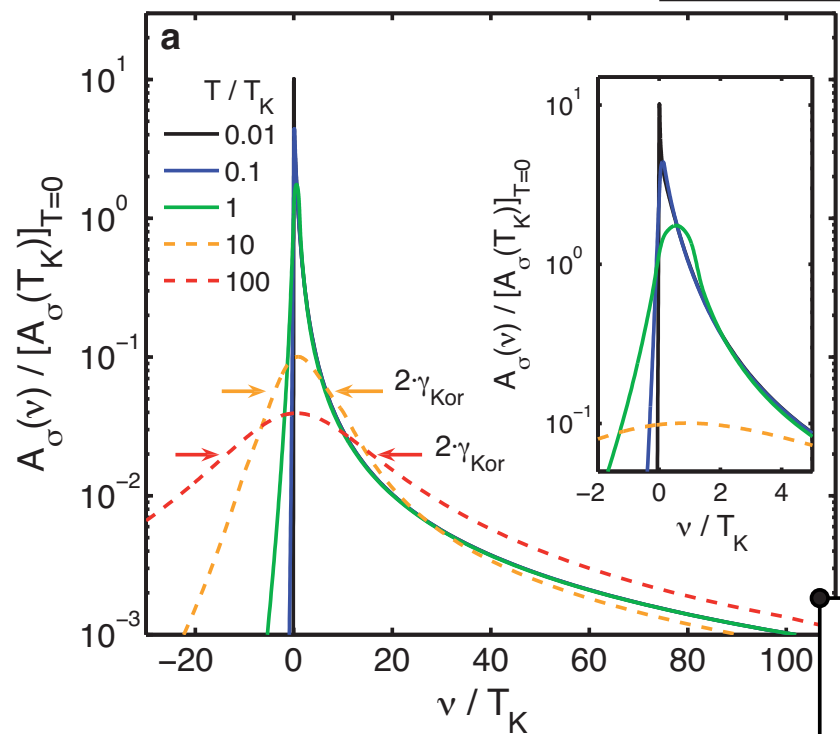
$\varepsilon_{e\sigma}^f$. Lower inset: Log-log plots of lineshapes for five different choices of $\varepsilon_{e\sigma}^f$, indicated by color-coded dashed lines in upper inset, and arrows in main panel. Main panel: When appropriately rescaled, the lineshapes collapse onto a universal curve in the LM-regime $T_K \lesssim \nu \lesssim |\varepsilon_{e\sigma}^f|$. In the SC-regime $T \lesssim \nu \lesssim T_K$, the curves do not collapse, since their exponents η_σ depend, via Δn_e , on $\varepsilon_{e\sigma}^f$.

Figure 4 Asymmetric magnetic-field dependence of lineshape at $T = 0$. **a**, Depending on whether the electron is photoexcited into the “lower” or “upper” of the Zeeman-split e-levels ($\sigma g_e B < 0$ or > 0 , solid or dashed lines, respectively), increasing $|B|$ causes the near-threshold divergence, $A_\sigma(\nu) \propto \nu^{-\eta_\sigma}$, to be either strengthened, or suppressed via the appearance of a peak at $\nu \simeq \sigma g_e B$, respectively. (The peak’s position is shown by the red line in the $\sigma g_e B$ - ν plane.) **b**, Universal dependence on $g_e B/T_K$ of the local moment m_e^f (dash-dotted line), and the corresponding prediction of Hopfield’s rule, Eq. (7), for the infrared exponents η_{lower} (solid line) and η_{upper} (dashed line) for $\sigma = +$. Symbols: η_σ -values extracted from the near-threshold $\nu^{-\eta_\sigma}$ divergence of $A_\sigma(\nu)$, for several magnetic fields and three values of Γ/U . Symbols and lines agree to within 1 %.

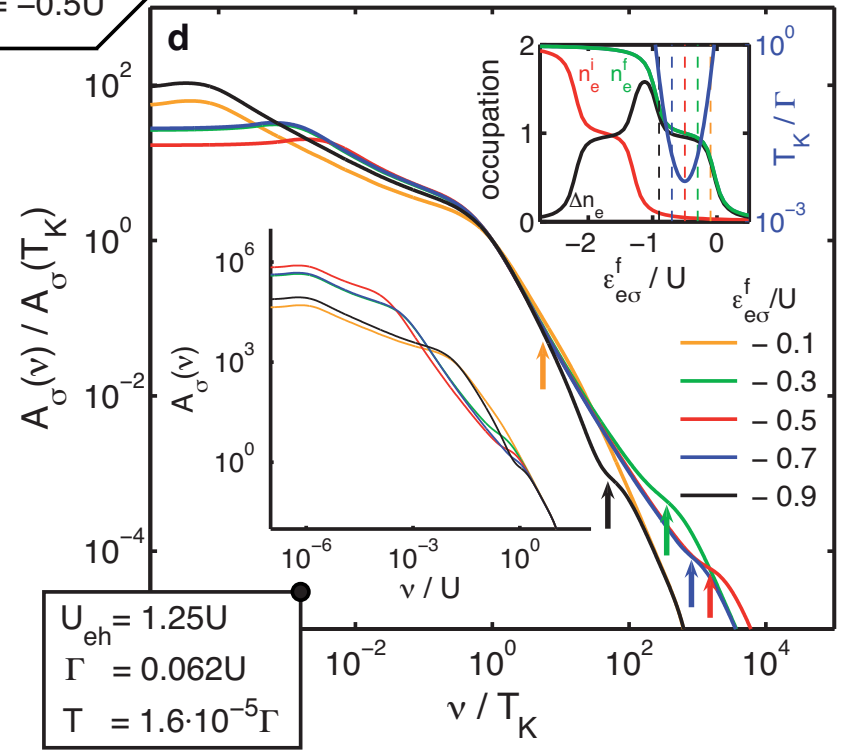
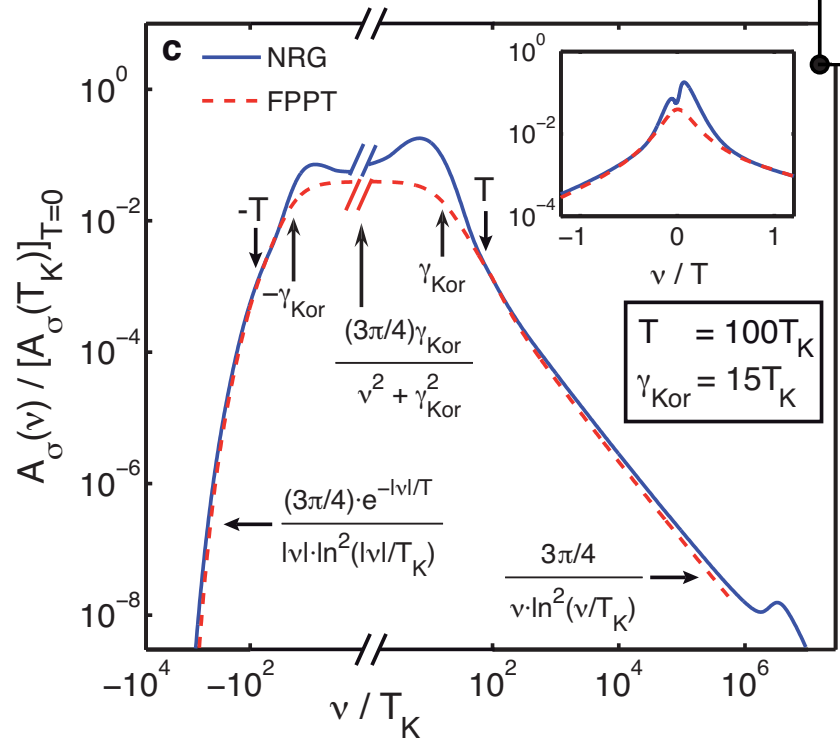
a**b****c**

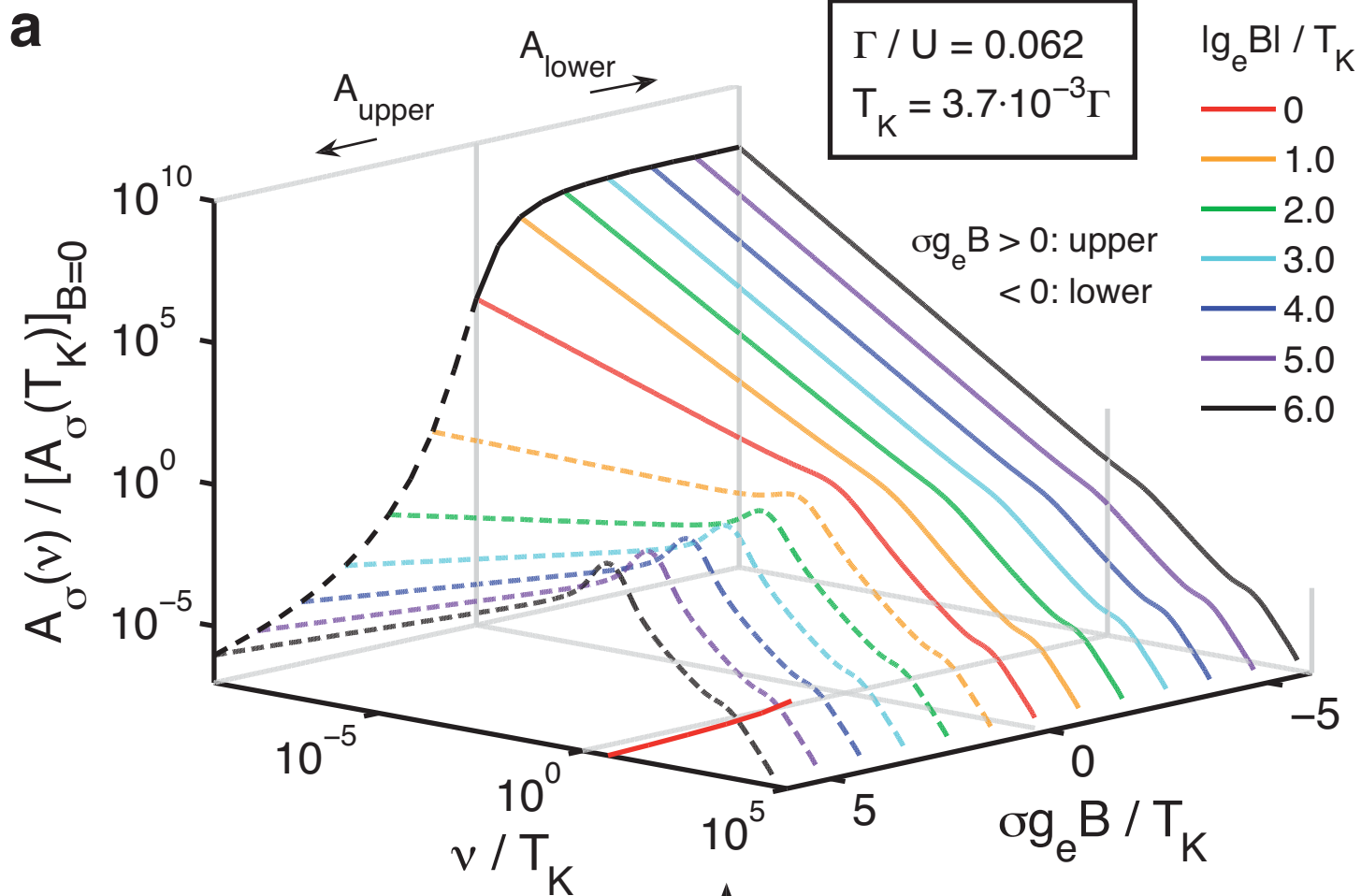


$U = 0.1D, B = 0$



$\Gamma = 0.03U$
 $T_K = 5.9 \cdot 10^{-6} \Gamma$
 $\epsilon_e^i = 0.75U$
 $\epsilon_e^f = -0.5U$





$U = 0.1D; \varepsilon_e^i = 0.75U; \varepsilon_e^f = -0.5U; T = 0$

