Many-Body Dynamics of Exciton Creation in a Quantum Dot by Optical Absorption: A Quantum Quench towards Kondo Correlations

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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 line shape and understood in terms of the three fixed points of the single-impurity Anderson model. At low temperatures the line shape 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.

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Many-body physics of quantum dots

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 the Kondo effect, in which QD and FR are bound into a spin singlet. It is interesting to ask how Kondo correlations set in after a quantum quench, i.e., a sudden change of the QD Hamiltonian, and corresponding predictions have been made in the context of transport experiments [1–4]. Optical transitions in quantum dots [5–7] offer an alternative arena for probing Kondo quenches: the creation of a bound electron-hole pair—an exciton—via photon absorption implies a sudden change in the local charge configuration. This induces a sudden switch-on of both a strong electron-hole attraction [6–8] and an exchange interaction between the bound electron and the FR. The subsequent dynamics is governed by energy scales that become ever lower with increasing time, leaving telltale signatures in the absorption and emission line shapes. For example, at low temperatures and small detunings relative to the threshold, the line shape has been predicted to show a gate-tunable power-law singularity [8]. Though optical signatures of Kondo correlations have not yet been experimentally observed, prospects for achieving this goal improved recently due to two key experimental advances [9,10].

Here we propose a realistic scenario for an optically induced quantum quench into a regime of strong Kondo correlations. A quantum dot tunnel coupled to a FR is prepared in an uncorrelated initial state [Fig. 1(a)]. Optical absorption of a photon creates an exciton, thereby inducing a quantum quench to a state conducive to Kondo correlations [Fig. 1(b)]. The subsequent emergence of spin correlations between the QD-electron and the FR, leading to a screened spin singlet, is imprinted on the optical absorption line shape [Fig. 1(c)]: its high-, intermediate-, and low-detuning behaviors are governed by the three fixed points of the single-impurity Anderson model (AM) [Fig. 1(d)]. We present detailed numerical and analytical results for the line shape as a function of temperature and magnetic field. At zero temperature we predict a tunable Anderson orthogonality catastrophe, since the difference in initial and final ground state phase shifts of FR electrons read off from the absorption line shape and understood in terms of the three fixed points of the single-impurity Anderson model (AM) [Fig. 1(d)]. We present detailed numerical and analytical results for the line shape as a function of temperature and magnetic field. At zero temperature we predict a tunable Anderson orthogonality catastrophe, since the difference in initial and final ground state phase shifts of FR electrons...
[indicated by wavy lines in Fig. 1(d)] can be tuned by magnetic field and gate voltage via their effects on the level occupancy.

Model.—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(a) and 1(b)]. In a gate voltage regime for which the QD is initially uncharged, a circularly polarized light beam (polarization \( \sigma \)) at a suitably chosen frequency \( \omega_L \) propagating along the \( z \) axis of the heterostructure will create a so-called neutral exciton [11] (\( X^0 \)), a bound electron-hole pair with well-defined spins \( \sigma \) and \( \bar{\sigma} = -\sigma \) (\( \in \{+,-\} \)) in the lowest available localized \( s \) orbitals of the QD’s conduction- and valence bands (to be called \( e \) and \( h \) levels, with creation operators \( e^{\dagger}_e \) and \( h^{\dagger}_h \), respectively). The QD-light interaction is described by \( H_L \propto (e^{\dagger}_h h^{\dagger}_e e^{-i\omega_L t} + \text{H.c.}) \), which we model the system before and after absorption by the initial and final Hamiltonian

\[
H^{\text{initial}} = H^{\text{final}} = H^{\text{final}} + H_c + H_t, \quad \text{where}
\]

\[
H_c = \sum_{\sigma} e^{\dagger}_e n_{e\sigma} + Un_e n_{e\sigma} + \delta_{af} e_{h\sigma} \quad (a = i,f) \quad (1)
\]

describes the QD, with Coulomb cost \( U \) for double occupancy of the \( e \) level, \( n_{e\sigma} = e^{\dagger}_e e_{\sigma} \), and hole energy \( e_{h\sigma} (>0, \text{on the order of the band gap}) \). The \( e \) level’s initial and final energies before and after absorption, \( e^{\dagger}_e \) \( (a = i, f) \), differ by the Coulomb attraction \( U_{eh} (>0) \) between the newly created electron-hole pair, which pulls the final \( e \) level downward, \( e^{\dagger}_e = e_{e\sigma} - \delta_{af} U_{eh} \) [Fig. 1(b)]. This stabilizes the excited electron against decay into the FR, provided that \( e_{e\sigma} \) lies below the FR’s Fermi level \( e_F \). Since \( H^{\text{final}} \neq H^{\text{initial}} \), absorption implements a quantum quench, which, as elaborated below, can be tuned by electric and magnetic fields. The term \( H_c = \sum_{\sigma} e^{\dagger}_e n_{e\sigma} + Un_e n_{e\sigma} + \delta_{af} e_{h\sigma} \) represents a noninteracting conduction band (the FR) with half-width \( D = 1/(2\rho) \) and constant density of states \( \rho \) per spin, with \( H_c = \sqrt{\pi \rho} \sum_{\sigma} (e^{\dagger}_e c_{\sigma} + \text{H.c.}) \), with \( c_{\sigma} = \sum_{\kappa} \epsilon_{\kappa}^{\sigma} \) 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, \( \epsilon_{e\sigma} = e_F + \frac{1}{2} \sigma g_B B \) (the Zeeman splitting of FR states can be neglected for our purposes [12]). The electron-hole pair created by photon absorption will additionally experience a weak but highly anisotropic intradot exchange interaction [12]. Its effects can be fully compensated by applying a magnetic field fine-tuned to a value, say \( B^0_{eh} \), that restores degeneracy of the \( e \) level’s two spin configurations [12]. Henceforth, \( B \) is understood to be measured relative to \( B^0_{eh} \). We set \( \mu_B = h = k_B = 1 \), give energies in units of \( D = 1 \) throughout, and assume \( T, B \ll \Gamma \ll U, U_{eh} \ll D \ll \epsilon_{h\sigma} \). The electron-hole recombination rate is assumed to be negligibly small compared to all other energy scales. We focus on the case, illustrated in Figs. 1(a) and 1(b), where the \( e \) level is essentially empty in the initial state and singly occupied in the ground state of the final Hamiltonian, \( n_{e\sigma} \approx 0 \) and \( n_{h\sigma} \approx 1 \). (Here \( n_{\sigma} = \sum_{\sigma} n_{e\sigma} \) and \( \bar{n}_{\sigma}^{\ddagger} = \langle n_{e\sigma} \rangle_{\sigma} \) is the thermal average of \( n_{e\sigma} \) with respect to \( H^f \).) This requires \( \epsilon_{e\sigma} \gg \Gamma, \) \( -U + \Gamma \ll \epsilon_{f\sigma} \ll -\Gamma \). The Kondo temperature associated with \( H^f \) is \( T_K = \sqrt{U/2\pi\epsilon(\epsilon_+ + U)/(2\Gamma)} \). If \( \epsilon_{e\sigma} = -U/2 \), so that \( \bar{n}_{e\sigma} = 1 \), then \( H^f \) represents the symmetric excitonic Anderson model, to be denoted by writing \( H^f = \text{SEAM} \).

Absorption line shape.—Absorption sets in once \( \omega_L \) exceeds a threshold frequency, \( \omega_{th} \). The line shape at temperature \( T \) and detuning \( \nu = \omega_L - \omega_{th} \) is, by the golden rule, proportional to the spectral function (see [13])

\[
A_{\sigma}(\nu) = 2\pi \sum_{m,n} p_{m,n} \int |m|^2 |\langle m|e_{\sigma}|n\rangle|^2 \delta(\omega_L - E_m^f, E_n^f). \quad (2)
\]

Here \( |m\rangle, |n\rangle \) and \( E_n^f, E_m^f \) are exact eigenstates and energies of \( H^f \), depicted schematically in Fig. 1(c), and \( p_{m,n} = e^{-E_n^f/T}/Zi \) the initial Boltzmann weights. The threshold frequency evidently is \( \omega_{th} = E_G^f - E_G^c \) (\( E_G^c \) is the ground state energy of \( H^f \)), which is on the order of \( \epsilon_{e\sigma} + \epsilon_{h\sigma} \) (up to corrections due to tunneling and correlations).

We calculated \( A_{\sigma}(\nu) \) using the Numerical Renormalization Group (NRG) [14], generalizing the approach of Refs. [8,15] to \( T \neq 0 \) by following Ref. [16]. The inset of Fig. 2 shows a typical result: As temperature is gradually reduced, an initially rather symmetric line shape becomes highly asymmetric, dramatically increasing in peak height as \( T \to 0 \). At \( T = 0 \), the line shape displays a threshold, vanishing for \( \nu < 0 \) and diverging as \( \nu \) tends to 0 from above. Figure 2 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 emerge in the regimes of “large”, “intermediate” or “small” detuning, labeled (for reasons discussed below) FO, LM and SC, respectively, (given here for \( H^f = \text{SEAM} \)):

\[
|\epsilon_{e\sigma}| \leq \nu \leq D: \quad A^{\text{FO}}_{\sigma}(\nu) = \frac{4\Gamma}{\nu^2} \theta(\nu - |\epsilon_{e\sigma}|); \quad (3a)
\]

\[
T_K \leq \nu \leq |\epsilon_{e\sigma}|: \quad A^{\text{LM}}_{\sigma}(\nu) = \frac{3\pi}{4\nu} \ln^{-2}(\nu/T_K); \quad (3b)
\]

\[
T \leq \nu \leq T_K: \quad A^{\text{SC}}_{\sigma}(\nu) \propto T_K^{-1}(\nu/T_K)^{-\eta_\sigma}. \quad (3c)
\]

The remarkable series of crossovers found above are symptomatic of three different regimes of charge and spin dynamics. They can be understood analytically using fixed-point perturbation theory (FPPT). To this end, note that at \( T = 0 \) the absorption line shape can be written as

\[
A_{\sigma}(\nu) = 2\text{Re} \int_0^\infty dt e^{i\epsilon_{e\sigma} t} G(t)e^{i\epsilon_{h\sigma} t} e^{-i\epsilon_{e\sigma} t} G(t), \quad (4)
\]

where \( H^a = H^f = E^0_B \) and \( \nu_+ = \nu + i\delta_+ \). Thus it directly probes the postquench dynamics, with initial state \( e^{\dagger}_f|G\rangle, \)
of a photogenerated e-electron coupled to a FR. Evidently, large, intermediate or small detuning, corresponding to ever longer time scales after absorption, probes excitations at successively smaller energy scales [see Fig. 1(c)], for which $\tilde{H}_f$ can be represented by expansions $H_f^\alpha + H_f^\beta$ around the three well-known fixed points [14] of the 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 a screened spin singlet, respectively, as illustrated in Fig. 1(d).

Large and intermediate detuning—perturbative regime.—For large detuning, probing the time interval $t \leq 1/|\epsilon_{e\sigma}|$ immediately after absorption, the $e$ level appears as a free, filled orbital perturbed by charge fluctuations, described by [14] the fixed-point Hamiltonian $H_{\text{FO}} = H_e + H_f^\alpha + \text{const}$ and the relevant perturbation $H_{\text{FO}}^r = H_{\text{FO}}$. Intermediate detuning probes the times $1/|\epsilon_{e\sigma}| \ll t \ll 1/T_K$ 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 by [14] $H_{\text{LM}}^r = H_e + \text{const}$ and the RG-relevant perturbation $H_{\text{LM}} = H_{\text{FO}} + H_f^\alpha + \text{const}$.

For $r = \text{FO}$ and LM, $A_\sigma(v)$ can be calculated using perturbation theory in $H_f^\alpha$. For $T = 0$, note that

$$A_\sigma(v) = -2 \text{Im} \langle G \rangle e_\sigma \frac{1}{v - H_f^\alpha} e_{\sigma'}^\dagger \langle G \rangle_i,$$

and

$$A_\sigma^r(v) \approx -2 \frac{v}{\nu^2} \text{Im} \langle G \rangle e_\sigma H_f^\alpha \frac{1}{v - H_f^\alpha} e_{\sigma'}^\dagger \langle G \rangle_i,$$

which reveals the relevant physics: Large detuning ($r = \text{FO}$) is described by the spectral function of the operator $H_e e_{\sigma'}^\dagger$; 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 $\tilde{s}_e \cdot \tilde{s}_e e_{\sigma'}^\dagger$, i.e., it probes spin fluctuations. Evaluating these spectral functions is elementary since $H_{\text{FO}}$ and $H_{\text{LM}}$ involve only free fermions. For $B = 0$ and $|\epsilon_{e\sigma}| = \frac{1}{2} U$, we readily recover Eqs. (3a) and (3b) (see [13]), which quantitatively agree with the NRG results of Fig. 2.—Though the latter was calculated for $H_f = \text{SEAM}$, Eq. (3b) holds more generally as long as $H_f^\alpha$ remains in the LM regime, with $n_l^\alpha \approx 1$; then $A_\sigma^r(v)$ depends on $\epsilon_{e\sigma}$, $U$ and $\Gamma$ only through their influence on $T_K$, and hence is a universal function of $v$ and $T_K$. The FPPT strategy for calculating FO and LM line shapes can readily be generalized to finite temperatures [12], using the methods of Ref. [17] (Section III.A) for the finite-T dynamic magnetic susceptibility [13]. For $|v| \ll |\epsilon_{e\sigma}|$ and $\max(|v|, T) \gg T_K$, we obtain

$$A_{\text{LM}}^\text{LM}(v) = \frac{3\pi}{4} \frac{v/T_K}{1 - e^{-v/T_K}} \gamma_{\text{Kor}}(v, T)/\pi,$$

where $\gamma_{\text{Kor}}(v, T) = \pi T/\ln^2[\max(|v|, T)/T_K]$ is the scale-dependent Korringa relaxation rate [17]. It is smaller than $T$ by a large logarithmic factor, implying a narrower and higher absorption peak than for thermal broadening.

Small detuning and Kondo-edge singularity—strong-coupling regime.—As $v$ is lowered through the bottom of the LM regime, $J(v)$ increases through unity into the strong-coupling regime, and $A_\sigma(v)$ monotonically crosses over to the SC regime. It was first studied for the present model (for $B = 0$) in Ref. [8], which found a power-law line shape of the form $(3c)$, characteristic of a Fermi edge singularity, with an exponent $\eta$ that followed Hopfield’s rule [18]. The power-law behavior reflects Anderson orthogonality [19,20]: it arises because the final ground state $|G_j\rangle$ that is reached in the long-time limit is characterized by a screened singlet. The singlet ground state induces different phase shifts [as indicated in Fig. 1(d) by wavy lines] for FR electrons than the unscreened initial state just after photon absorption, $e_{\sigma'}^\dagger |G_j\rangle$, and hence is orthogonal to the latter. It is straightforward to generalize
Since these cases yield fully asymmetric changes in local components, the lower or upper of the Zeeman-split pair (σgB ≠ 0 for photo-excitation into the lower level, since subsequently the e-level spin need not adjust at all. In contrast, it is maximal (Δn′_e,σ = 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 longtime value. It follows, remarkably, that a magnetic field tunes the strength of Anderson orthogonality, implying a dramatic asymmetry for the evolution of the line shape A_e(ν) ≈ ν^−ne with increasing |B| [Fig. 3(a)].

Conclusions.—We have shown that optical absorption in a single quantum dot can implement a quantum quench in an experimentally accessible solid-state system that allows the emergence of Kondo correlations and Anderson orthogonality to be studied in a tunable setting.

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FIG. 3 (color online). Asymmetric magnetic-field dependence of the line shape for H' = SEAM and T = 0. (a) Depending on whether the electron is photoexcited into the lower or upper of the Zeeman-split e levels (σgB < 0 or >0, solid or dashed lines, respectively), increasing |B| causes the near-threshold divergence, A_e(ν) ≈ ν^−ne, to be either strengthened, or suppressed via the appearance of a peak at ν ≈ σgB, respectively. (The peak’s position is shown by the red line in the σg-B-ν plane.) (b) Universal dependence on g_eB/T_K of the local moment m_e^i (dash-dotted line), and the corresponding prediction of Hopfield’s rule, Eq. (8), for the infrared exponents η_lower (solid line) and η_upper (dashed line) for σ = ±1. Symbols: η_± values extracted from the near-threshold ν^−η_± divergence of A_e(ν). Symbols and lines agree to within 1%.

The arguments of Refs. [8,18] to the case of B ≠ 0 (see [13]). One readily finds the generalized Hopfield rule

\[ η_σ = 1 - \sum_{σ'} (Δn′_{σ,σ'})^2, \quad Δn′_{σ,σ'} = δ_{σ,σ'} - Δn_{σ,σ'}, \]  

\[ \Delta n_{σ,σ'} = \frac{1}{2} \left( \frac{2m_e^f - 2m_e^i}{J - 1} \right), \]

where the final magnetization m_e^f is a universal function of g_eB/T_K. The exponents η_σ then are universal functions of g_eB/T_K, with simple limits for small and large fields (see Fig. 3(b)); η_σ → 1/2 for [g_eB] ≪ T_K, while η_lower/upper → ±1 for [g_eB] ≫ T_K. Here the subscript “lower” or “upper” distinguishes whether the spin-σ electron is photoexcited into the lower or upper of the Zeeman-split pair (σgB < 0 or >0, respectively). The sign difference ≥ 1 for η_σ arises since these cases yield fully asymmetric changes in local charge: Δn_e,lower → 1 while Δn_e,upper → 0. As a result, Anderson orthogonality [19] is completely absent (Δn_e,σ = 0) for photo-excitation into the lower level, since subsequently the e-level spin need not adjust at all. In contrast, it is maximal (Δn_e,σ = 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 longtime value. It follows, remarkably, that a magnetic field tunes the strength of Anderson orthogonality, implying a dramatic asymmetry for the evolution of the line shape A_e(ν) ≈ ν^−ne with increasing |B| [Fig. 3(a)].

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