

EPAPS: Supplementary information
for “Many-Body Dynamics of Exciton Creation in a Quantum
Dot by Optical Absorption:
A Quantum Quench towards Kondo Correlations”

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We provide below some intermediate steps for the derivation of the main equations of the main text. For clarity, information contained in the main text is typeset in blue.

Spectral function

The origin of Eq. (2) for the absorption rate can be understood as follows [1]. We begin with a Hamiltonian slightly more general than those of the main text, in that it includes the hole degree of freedom: $H = H_{\text{eh}} + H_{\text{c}} + H_{\text{t}}$, where H_{c} and H_{t} are given in the main text, and

$$H_{\text{eh}} = \sum_{\sigma} (\varepsilon_{\text{e}\sigma} n_{\text{e}\sigma} + \varepsilon_{\text{h}\sigma} n_{\text{h}\sigma}) + U n_{\text{e}\uparrow} n_{\text{e}\downarrow} - \sum_{\sigma\sigma'} U_{\text{eh}} n_{\text{e}\sigma} n_{\text{h}\sigma'} \quad (\text{S1})$$

describes the QD, with e-level charging energy $U(>0)$, e-h Coulomb attraction $U_{\text{eh}}(>0)$, $n_{\text{e}\sigma} = e_{\sigma}^{\dagger} e_{\sigma}$, $n_{\text{h}\sigma} = h_{\sigma}^{\dagger} h_{\sigma}$. The hole energy $\varepsilon_{\text{h}\bar{\sigma}}$ (>0) is on the order of the band gap.

The QD-light interaction is described by $H_{\text{L}} \propto (e_{\sigma}^{\dagger} h_{\bar{\sigma}}^{\dagger} e^{-i\omega_{\text{L}}t} + \text{h.c.})$. Absorption sets in once ω_{L} exceeds a threshold frequency, say ω_{th} . According to Fermi's golden rule, treating H_{L} as harmonic perturbation, the absorption lineshape at temperature T and detuning $\nu = \omega_{\text{L}} - \omega_{\text{th}}$ is proportional to

$$A_{\sigma}(\nu) = 2\pi \sum_{mm'} \rho_m |\langle m' | e_{\sigma}^{\dagger} h_{\bar{\sigma}}^{\dagger} | m \rangle|^2 \delta(\omega_{\text{L}} - E_{m'} + E_m), \quad (\text{S2})$$

where the ρ_m are Boltzmann weights. Noting that the dynamics of the optically created hole is trivial, $[n_{\text{h}\sigma}, H] = 0$, we can write all initial states $|m\rangle$ as $|m\rangle_{\text{i}} \otimes |0\rangle_{\text{h}}$ and all final states $|m'\rangle$ in the form $|m'\rangle_{\text{f}} \otimes |\bar{\sigma}\rangle_{\text{h}}$, thus arriving at Eq. (2) from the main text:

$$A_{\sigma}(\nu) = 2\pi \sum_{mm'} \rho_m^{\text{i}} |\langle m' | e_{\sigma}^{\dagger} | m \rangle_{\text{i}}|^2 \delta(\omega_{\text{L}} - E_{m'}^{\text{f}} + E_m^{\text{i}}). \quad (\text{S3})$$

Here $|m\rangle_a$ ($a = \text{i, f}$) are the many-body eigenstates of the effective initial and final Hamiltonians, $H^{\text{i}} = {}_{\text{h}}\langle 0 | H | 0 \rangle_{\text{h}}$ and $H^{\text{f}} = {}_{\text{h}}\langle \bar{\sigma} | H | \bar{\sigma} \rangle_{\text{h}}$, that only include the QD and FR electronic

degrees of freedom. They are given by $H^{i/f} = H_e^{i/f} + H_c + H_t$, with $H_e^{i/f}$ given by Eq. (1):

$$H_e^a = \sum_{\sigma} \varepsilon_{e\sigma}^a n_{e\sigma} + U n_{e\uparrow} n_{e\downarrow} + \delta_{af} \varepsilon_{h\sigma} \quad (a = i, f), \quad \text{with} \quad \varepsilon_{e\sigma}^a = \varepsilon_{e\sigma} - \delta_{af} U_{eh}. \quad (\text{S4})$$

To bring the spectral function into a resolvent form suitable for fixed point perturbation theory (FPPT), we use Dirac's identity

$$\delta(\omega_L - E_{m'}^f + E_m^i) = -\frac{1}{\pi} \text{Im} \left[\frac{1}{\omega_L - E_{m'}^f + E_m^i + i0^+} \right], \quad (\text{S5})$$

to rewrite Eq. (S3) as follows:

$$A_{\sigma}(\nu) = -2 \text{Im} \left[\sum_{mm'} \rho_m^i \langle m | e_{\sigma} \frac{1}{\omega_L - E_{m'}^f + E_m^i + i0^+} | m' \rangle_f \langle m' | e_{\sigma}^{\dagger} | m \rangle_i \right]. \quad (\text{S6})$$

$$= -2 \text{Im} \left[\sum_m \rho_m^i \langle m | e_{\sigma} \frac{1}{\omega_L - H^f + E_m^i + i0^+} e_{\sigma}^{\dagger} | m \rangle_i \right]. \quad (\text{S7})$$

For the last step, we replaced $E_{m'}^f$ by H^f and used $\sum_{m'} |m'\rangle_f \langle m'| = 1$.

Zero temperature:

At $T = 0$, the initial density matrix is $|G\rangle_i \langle G|$. With the definitions introduced in the text, $\tilde{H}^a = H^a - E_G^a$, $\nu = \omega_L - E_G^f + E_G^i$ and $\nu_+ = \nu + i0_+$, Eq. (S7) reduces to Eq. (5) of the main text:

$$A_{\sigma}(\nu) = -2 \text{Im} \langle G | e_{\sigma} \frac{1}{\nu_+ - \tilde{H}^f} e_{\sigma}^{\dagger} | G \rangle_i. \quad (\text{S8})$$

For the parameters considered in the main text, the ground state of H^i can be approximated by a free Fermi sea: $|G\rangle_i \simeq \prod_{\varepsilon_{k\sigma} < \varepsilon_F} c_{k\sigma}^{\dagger} |\text{Vac}\rangle$. Next comes the key step of FPPT: replace $\tilde{H}^f \rightarrow H_r^* + H_r'$ and do a perturbation expansion in H_r' of the resolvent

$$\frac{1}{\nu_+ - \tilde{H}^f} = \frac{1}{\nu_+ - H_r^*} + \frac{1}{\nu_+ - H_r^*} \hat{\mathcal{T}}_r \frac{1}{\nu_+ - H_r^*} \quad (\text{S9})$$

with the T-matrix given by

$$\hat{\mathcal{T}}_r = H_r' + H_r' \frac{1}{\nu_+ - H_r^*} H_r' + \dots \quad (\text{S10})$$

According to the main text, the free orbital (FO) regime is described by

$$H_{\text{FO}}^* = H_c + H_e^f - E_{\text{FO}} = \sum_{k\sigma} \varepsilon_{k\sigma} c_{k\sigma}^{\dagger} c_{k\sigma} + \sum_{\sigma} \varepsilon_{e\sigma}^f n_{e\sigma} + U n_{e\uparrow} n_{e\downarrow} - E_{\text{FO}}, \quad (\text{S11a})$$

perturbed by

$$H_{\text{FO}}' = H_t = \sqrt{\Gamma/\pi\rho} \sum_{\sigma} (e_{\sigma}^{\dagger} c_{\sigma} + \text{h.c.}), \quad (\text{S11b})$$

and the local moment (LM) regime by

$$H_{\text{LM}}^* = H_{\text{c}} - E_{\text{LM}} = \sum_{k\sigma} \varepsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} - E_{\text{LM}} , \quad (\text{S12a})$$

perturbed by

$$H'_{\text{LM}} = \frac{J(\nu)}{\rho} \vec{s}_{\text{e}} \cdot \vec{s}_{\text{c}} , \quad (\text{S12b})$$

with e-level and conduction band spin operators given by $\vec{s}_j = \frac{1}{2} \sum_{\sigma\sigma'} j_\sigma^\dagger \vec{\tau}_{\sigma\sigma'} j_{\sigma'}$ (for $j = e, c$). The subtracted constants E_{FO} and E_{LM} correspond to the subtraction of E_{G}^{f} in the definition of $\bar{H}^{\text{f}} = H^{\text{f}} - E_{\text{G}}^{\text{f}} \rightarrow H_r^* + H'_r$ (see the main text after Eq. (4)); they ensure that the ground state of H_r^* has eigenvalue 0. For both $r = \text{FO}$ and LM , this ground state is given by $|\sigma\rangle \equiv e_\sigma^\dagger |\text{G}_i\rangle$.

Now, when inserted into Eq. (S8), the first term in Eq. (S9) gives a $\delta(\nu)$ not relevant for the regime $\nu \gtrsim T_K$ that we are focusing on. The second term gives $-2\text{Im}\langle\sigma|\widehat{\mathcal{T}}_r|\sigma\rangle/\nu^2 = \text{Im}[\mathcal{T}_r^{(1)} + \mathcal{T}_r^{(2)} + \dots]/\nu^2$. For both $r = \text{FO}$ and LM , $\mathcal{T}_r^{(1)} = \langle\sigma|H'_r|\sigma\rangle = 0$ (for $B = 0$). Thus, to lowest non-zero order, we obtain Eq. (6) of the main text, namely

$$A_\sigma^r(\nu) = -2\text{Im}[\mathcal{T}_r^{(2)}]/\nu^2, \quad \mathcal{T}_r^{(2)} = \langle\sigma|H'_r \frac{1}{\nu_+ - H_r^*} H'_r|\sigma\rangle. \quad (\text{S13})$$

$\mathcal{T}_r^{(2)}$ can be evaluated straightforwardly using Wick's theorem, since $H'_r|\sigma\rangle$ produces a sum of uncorrelated free-fermion states, all of which are eigenstates of $\frac{1}{\nu_+ - H_r^*}$. For the free orbital regime, we obtain

$$\mathcal{T}_{\text{FO}}^{(2)} = \left(\sqrt{\Gamma/\pi\rho}\right)^2 \sum_{ks, k's'} \langle\sigma| \left(c_{ks}^\dagger e_s + \text{h.c.}\right) \frac{1}{\nu_+ - H_{\text{FO}}^*} \left(c_{k's'}^\dagger e_{s'} + \text{h.c.}\right) |\sigma\rangle \quad (\text{S14})$$

$$= \frac{\Gamma}{\pi\rho} \sum_k \left[\frac{(1 - f(\varepsilon_k))}{\nu_+ - (\varepsilon_k - \varepsilon_{\text{e}}^{\text{f}})} + \frac{f(\varepsilon_k)}{\nu_+ - (-\varepsilon_k + \varepsilon_{\text{e}}^{\text{f}} + U)} \right] , \quad (\text{S15})$$

where $f(\varepsilon) = \theta(-\varepsilon)$ stands for the Fermi function at zero temperature. Inserting this into Eq. (S13) we obtain (for $D \gg |\nu|$):

$$A_\sigma^{\text{FO}}(\nu) = \frac{2\Gamma}{\nu^2} \left[\theta(\nu - |\varepsilon_{\text{e}\sigma}^{\text{f}}|) + \theta(\nu - (\varepsilon_{\text{e}\sigma}^{\text{f}} + U)) \right] . \quad (\text{S16})$$

For the case of $H^{\text{f}} = \text{SEAM}$ (with $\varepsilon_{\text{e}}^{\text{f}} = -U/2$) considered in the main text, this reduces to Eq. (3a):

$$A_\sigma^{\text{FO}}(\nu) = \frac{4\Gamma}{\nu^2} \theta(\nu - |\varepsilon_{\text{e}\sigma}^{\text{f}}|) . \quad (\text{S17})$$

The calculation for the local moment regime, $T_K \ll |\nu| \ll \min[|\varepsilon_{\text{e}\sigma}^{\text{f}}|, \varepsilon_{\text{e}\sigma}^{\text{f}} + U]$, is analogous.

Writing $\vec{s}_e \cdot \vec{s}_c = \frac{1}{2}(s_e^\sigma s_c^{\bar{\sigma}} + s_e^{\bar{\sigma}} s_c^\sigma) + s_e^z s_c^z$, we obtain

$$\mathcal{T}_{\text{LM}}^{(2)} = \left(\frac{J(\nu)}{\rho} \right)^2 \langle \sigma | \vec{s}_e \cdot \vec{s}_c \frac{1}{\nu_+ - H_{\text{LM}}^*} \vec{s}_e \cdot \vec{s}_c | \sigma \rangle \quad (\text{S18a})$$

$$= \frac{1}{4} \left(\frac{J(\nu)}{\rho} \right)^2 \left[\langle G_i | s_c^z \frac{1}{\nu_+ - H_{\text{LM}}^*} s_c^z | G_i \rangle + \langle G_i | s_c^{\bar{\sigma}} \frac{1}{\nu_+ - H_{\text{LM}}^*} s_c^\sigma | G_i \rangle \right] \quad (\text{S18b})$$

$$= \frac{3}{8} \left(\frac{J(\nu)}{\rho} \right)^2 \sum_{kq} \frac{f(\varepsilon_k)(1 - f(\varepsilon_q))}{\nu_+ - \varepsilon_q + \varepsilon_k}. \quad (\text{S18c})$$

Inserting Eq. (S18c) into Eq. (S13), with $J(\nu) = \ln^{-1}[\nu/T_K]$, we recover Eq. (3b):

$$A_\sigma^{\text{LM}} = \frac{3\pi}{4\nu} \frac{1}{\ln^2(\nu/T_K)}. \quad (\text{S19})$$

Nonzero temperature

For $T \neq 0$, the calculations are analogous, with only minor changes: The FPPT expansion in powers of H_r' is performed on Eq. (S7) (instead of Eq. (S8)), the Fermi occupation function is $f(\varepsilon) = 1/[e^{\varepsilon/T} + 1]$, and for the local moment regime the exchange coupling now takes the form $J(\nu) = \ln^{-1}[\max(|\nu|, T)/T_K]$. We consider only the local moment regime, with $|\nu| \ll \min[|\varepsilon_{e\sigma}^f|, \varepsilon_{e\sigma}^f + U]$ and $\max[|\nu|, T] \gg T_K$. Assuming (as before) that the initial level position lies so far above the Fermi surface ($\varepsilon_{e\sigma}^i \gg \Gamma$) that the initial density matrix contains no correlations between e-level and Fermi reservoir, we again arrive at Eq. (S18c), which now yields

$$A_\sigma^{\text{LM}} = \frac{3\pi}{4\nu} \frac{1}{1 - e^{-\nu/T}} \frac{1}{\ln^2[\max(|\nu|, T)/T_K]}. \quad (\text{S20})$$

For large positive detuning, $\nu \gg T$, we recover Eq. (S19), while the line-shape at large negative detuning, $\nu \ll -T$, is suppressed by an extra factor $e^{-|\nu|/T}$.

In the limit of small detuning, $|\nu| \ll T$, Eq. (S20) reduces to

$$A_\sigma^{\text{LM}}(\nu) = \frac{3\pi}{4\nu^2} \frac{T}{\ln^2[T/T_K]}. \quad (\text{S21})$$

The apparent ν^{-2} divergence indicates that in this limit, the expansion (S10) of $\mathcal{T}_{\text{LM}}$ can not be truncated at second order, as done above, but must be summed to all orders. Instead of doing this explicitly, one may use methods which were applied to treat the dynamic magnetic susceptibility at finite temperature in Ref. [2]. These yield

$$A_\sigma^{\text{LM}}(\nu) = \frac{3\pi}{4} \frac{\nu/T}{1 - e^{-\nu/T}} \frac{\gamma_{\text{Kor}}(\nu, T)/\pi}{\nu^2 + \gamma_{\text{Kor}}^2(\nu, T)}, \quad (\text{S22})$$

(Eq. (7) of the main text), which contains a Lorentzian factor involving a frequency-dependent Korringa relaxation time,

$$\gamma_{\text{Kor}}(\nu, T) = \frac{\pi T}{\ln^2[\max(|\nu|, T)/T_K]}. \quad (\text{S23})$$

For $|\nu| \gg \gamma_{\text{Kor}}(\nu, T)$, Eq. (S22) reproduces Eq. (S20). For $|\nu| \ll T$ (but $T \gg T_K$), Eq. (S22) reduces to a pure Lorentzian

$$A_{\sigma}^{\text{LM}}(\nu) = \frac{3\pi}{4} \frac{\gamma_{\text{Kor}}/\pi}{\nu^2 + \gamma_{\text{Kor}}^2}, \quad (|\nu| \ll T) \quad (\text{S24})$$

of width $\gamma_{\text{Kor}} \simeq \pi T / \ln^2[T/T_K]$ (which is $\ll T$). This represents the properly regularized version of Eq. (S21), to which it reduces for $\gamma_{\text{Kor}} \ll |\nu| \ll T$.

Generalized Hopfield rule

The generalized Hopfield rule that holds when $\nu \ll T_K$ and arbitrary B is stated in Eq. (8) of the main text can be found as follows, using arguments similar to those in Refs. [1, 3]: First, write the $T = 0$ spectral function of Eq. (4) as

$$A_{\sigma}(\nu) = 2 \text{Re} \int_0^{\infty} dt e^{it\nu} \langle \psi_0 | \psi_t \rangle, \quad (\text{S25})$$

where $|\psi_0\rangle = e_{\sigma}^{\dagger} |G_i\rangle$ is the state just after photon absorption (at $t = 0^+$) and $|\psi_t\rangle = e^{-i\hat{H}t} |\psi_0\rangle$ its time-evolved version. In the $t \rightarrow \infty$ limit (relevant for $\nu \rightarrow 0$), the dynamics is governed by the final ground state, $|G_f\rangle$, characterized by a screened spin singlet. Once the latter begins to dominate (for $t \gtrsim 1/T_K$), the FR experiences the QD as a site of pure potential scattering (no spin-flips), just as at $t = 0^+$, but with changed strength. The adjustment of the FR to this changed potential (via changes in the scattering phase shifts of its single-particle wave-functions) causes an increasing Anderson orthogonality [4] between $|\psi_t\rangle$ and $|\psi_0\rangle$: their overlap decays as $\langle \psi_0 | \psi_t \rangle \sim t^{-\sum_{\sigma'} (\Delta n'_{e\sigma'})^2}$ [5], where, by Friedel's sum rule [7, 6], $\Delta n'_{e\sigma'} = \delta_{\sigma\sigma'} - \Delta n_{e\sigma'}$ is the *displaced charge* (of spin σ'), in units of e , that flows from the scattering site to infinity as $e_{\sigma}^{\dagger} |G_i\rangle$ evolves to $|G_f\rangle$, with $\Delta n_{e\sigma'} = \bar{n}_{e\sigma'}^f - \bar{n}_{e\sigma'}^i$ the local occupation difference between $|G_f\rangle$ and $|G_i\rangle$. Fourier-transforming $\langle \psi_0 | \psi_t \rangle$ according to Eq. (S25) yields the powerlaw-decay of Eq. (3c), with exponent

$$\eta_{\sigma} = 1 - \sum_{\sigma'} (\Delta n'_{e\sigma'})^2. \quad (\text{S26})$$

This is the generalized Hopfield rule, Eq. (8).

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