Spin Correlations in Nonlinear Optical Response: Light-Induced Kondo Effect

T. V. Shahbazyan and I. E. Perakis
Department of Physics and Astronomy, Vanderbilt University, Nashville, Tennessee 37235

M. E. Raikh
Department of Physics, University of Utah, Salt Lake City, Utah 84112
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We study the role of spin correlations in nonlinear absorption due to transitions from a deep impurity level to states above a Fermi sea. We demonstrate that the Hubbard repulsion between two electrons at the impurity leads to a logarithmic divergence in $\chi^{(3)}$ at the absorption threshold. This divergence is a manifestation of the Kondo physics in the nonlinear optical response of Fermi sea systems. We also show that, for off-resonant pump excitation, the pump-probe spectrum exhibits a narrow peak below the linear absorption onset. Remarkably, the light-induced Kondo temperature, which governs the shape of the Kondo-absorption spectrum, can be *tuned* by varying the intensity and frequency of the pump.

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There are two prominent many-body effects in the linear absorption spectrum due to optical transitions from a localized impurity level to the continuum of states above a Fermi sea (FS). First is the Mahan singularity due to the attractive interaction between the FS and the localized hole. Second is the Anderson orthogonality catastrophe due to the readjustment of the FS density profile during the optical transition. Both effects have long become textbook material [1]. The role of many-body correlations in the *nonlinear* optical response has been investigated only during the past decade [2]. Recently, there has been a growing interest in the coherent ultrafast dynamics of the FS systems at low temperatures [3–8].

In this paper, we suggest a new many-body effect in the nonlinear absorption of a FS system with a deep impurity level. This effect originates from the *spin* correlations between the photoexcited and the FS electrons. We note that a number of different intermediate processes contribute to the third-order optical susceptibility $\chi^{(3)}$ [9]. It is crucial that, in the system under study, some of the intermediate states involve a doubly occupied impurity level. For example, the optical field can first cause a transition of a FS electron to the singly occupied impurity level, which thus becomes doubly occupied, and then excite both electrons from the impurity level to the conduction band. This is illustrated in Fig. 1(a). Importantly, while on the impurity, the two electrons experience a Hubbard repulsion. Our main observation is that such a repulsion gives rise to an anomaly in $\chi^{(3)}$. The origin of this anomaly is intimately related to the Kondo effect.

To be specific, we restrict ourselves to pump-probe spectroscopy, where a strong pump and a weak probe optical field are applied to the system, and the optical polarization along the probe direction is measured. We only consider near-threshold absorption at zero temperature and assume that the pump frequency is tuned below the onset of optical transitions from the impurity level so that dephasing processes due to electron-electron and electron-phonon interactions are suppressed. Under such excitation conditions, the following Hamiltonian describes the system: $H_{\rm tot} = H + H_1(t) + H_2(t)$, where

$$H = \sum_{\mathbf{k}\sigma} \varepsilon_k c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} + \varepsilon_d \sum_\sigma d^\dagger_\sigma d_\sigma + \frac{U}{2} \sum_{\sigma \neq \sigma'} \hat{n}_\sigma \hat{n}_{\sigma'}, \tag{1}$$

is the Hamiltonian in the absence of optical fields. Here $c^\dagger_{\mathbf{k}\sigma}$ and $d^\dagger_\sigma$ are conduction and localized electron creation operators, respectively ($\hat{n}_\sigma = d^\dagger_\sigma d_\sigma$), ε_k and ε_d are the corresponding energies, and U is the Hubbard interaction (all energies are measured from the Fermi level). The coupling to the optical fields is described by the Hamiltonian $H_i(t) = -M_i(t)\hat{T}^\dagger + \text{H.c.}$, where $\hat{T}^\dagger = \sum_{\mathbf{k}\sigma} c^\dagger_{\mathbf{k}\sigma} d_\sigma$ ($i = 1, 2$ denotes the probe and pump, respectively) with $M_i(t) = e^{i\mathbf{k}_i\cdot\mathbf{r} - i\omega_i t}\mu \mathcal{E}_i(t)$. Here $\mathcal{E}_i(t)$, $\mathbf{k}_i$, and ω_i are

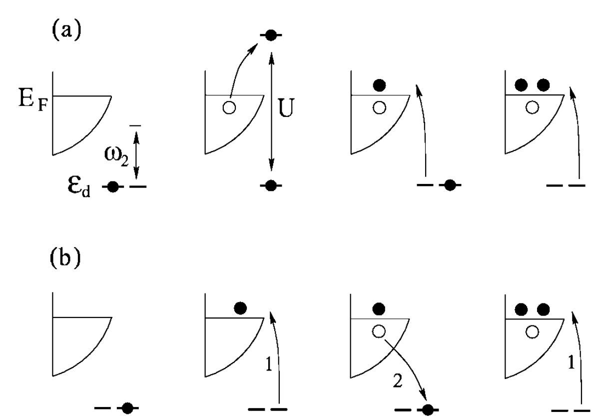

FIG. 1. Intermediate processes contributing to $\chi^{(3)}$. (a) Intermediate state with doubly occupied impurity. (b) Large U limit: *two* transition channels are available from states *below* the FS to the *empty* impurity, but only *one* channel from the *singly occupied* impurity to states *above* the FS.

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the pump-probe electric field amplitude, direction, and central frequency, respectively, and μ is the dipole matrix element. The pump-probe polarization is obtained by expanding the optical polarization, $\mu\langle\hat{T}\rangle$, to the first order in H_1 and keeping the terms propagating in the probe direction [9]:

$$P(t) = i\mu\int_{-\infty}^{t} dt'\, M_1(t')\,[\langle\Phi(t)|\hat{T}\mathcal{K}(t,t')\hat{T}^\dagger|\Phi(t')\rangle - \langle\Phi(t')|\hat{T}^\dagger\mathcal{K}(t',t)\hat{T}|\Phi(t)\rangle], \tag{2}$$

where $\mathcal{K}(t,t')$ is the evolution operator for the Hamiltonian $H + H_2(t)$ and the state $|\Phi(t)\rangle$ satisfies the Schrödinger equation $i\partial_t|\Phi(t)\rangle = [H + H_2(t)]|\Phi(t)\rangle$.

The third-order polarization is obtained by expanding $\mathcal{K}(t,t')$ and $|\Phi(t)\rangle$ up to the second order in H_2. Below we consider sufficiently large values of U so that, in the absence of optical fields, the ground state of H, $|\Omega_0\rangle$, represents a *singly occupied* impurity and full FS. For large U, the doubly occupied impurity states are energetically unfavorable and can be excluded from the expansion of the polarization (2) with respect to H_2. The third-order pump-probe polarization then takes the form $P^{(3)}(t) = e^{i\mathbf{k}_1\cdot\mathbf{r}-i\omega_1 t}\tilde{P}^{(3)}$ with

$$\tilde{P}^{(3)} = i\mu^4\int_{-\infty}^{t} dt'\,\mathcal{E}_1(t')e^{i\omega_1(t-t')} \times [Q_1(t,t') + Q_1^*(t',t) + Q_2(t,t') + Q_3(t,t')], \tag{3}$$

where

$$\begin{aligned} Q_1(t,t') &= -\int_{-\infty}^{t'} dt_1\int_{-\infty}^{t_1} dt_2\, f(t_1,t_2)F(t,t',t_1,t_2),\\ Q_2(t,t') &= -\int_{t'}^{t} dt_2\int_{t'}^{t_2} dt_1\, f(t_1,t_2)F(t,t_2,t_1,t'),\\ Q_3(t,t') &= -\int_{-\infty}^{t'} dt_1\int_{-\infty}^{t} dt_2\, f(t_1,t_2)F(t_1,t',t,t_2). \end{aligned} \tag{4}$$

Here we denoted $f(t_1,t_2) = \mathcal{E}_2(t_1)\mathcal{E}_2(t_2)e^{i\omega_2(t_1-t_2)}$, and

$$\begin{aligned} F(t,t',t_1,t_2) &= \langle\Omega_0|\hat{T}e^{-iH(t-t')}\hat{T}^\dagger e^{-iH(t'-t_1)}\hat{T}e^{-iH(t_1-t_2)}\hat{T}^\dagger|\Omega_0\rangle\\ &= \sum_{\mathbf{pqk'k}\lambda s\sigma'\sigma} A^{\lambda s\sigma'\sigma}_{\mathbf{pqk'k}}\, e^{-i(\varepsilon_p-\varepsilon_d)(t-t')-i(\varepsilon_k-\varepsilon_{k'})(t'-t_1)-i(\varepsilon_k-\varepsilon_d)(t_1-t_2)}, \end{aligned} \tag{5}$$

$$\begin{aligned} A^{\lambda s\sigma'\sigma}_{\mathbf{pqk'k}} &= \langle\Omega_0|d_\lambda^\dagger c_{\mathbf{p}\lambda}c_{\mathbf{q}s}^\dagger d_s d_{\sigma'}^\dagger c_{\mathbf{k'}\sigma'}c_{\mathbf{k}\sigma}^\dagger d_\sigma|\Omega_0\rangle\\ &= \delta_{\lambda\sigma}\delta_{s\sigma'}n_\sigma(1-n_p)\\ &\quad\times[\delta_{\mathbf{pk}}\delta_{\mathbf{qk'}}n_q + \delta_{\sigma\sigma'}\delta_{\mathbf{pq}}\delta_{\mathbf{kk'}}(1-n_k)], \end{aligned} \tag{6}$$

with $n_\sigma = \langle\Omega_0|d_\sigma^\dagger d_\sigma|\Omega_0\rangle$ and $n_k = \langle\Omega_0|c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma}|\Omega_0\rangle$ (the impurity occupation number here is $n_d = \sum_\sigma n_\sigma = 1$). For monochromatic optical fields, $\mathcal{E}_i(t) = \mathcal{E}_i$, the time integrals can be explicitly evaluated. After a lengthy but straightforward calculation, the third-order polarization (3) takes the form $\tilde{P}^{(3)} = \tilde{P}_0^{(3)} + \tilde{P}_K^{(3)}$ with

$$\tilde{P}_0^{(3)} = \mu^4\mathcal{E}_1\mathcal{E}_2^2\sum_{\mathbf{pq}}\frac{(1-n_p)}{\varepsilon_p-\varepsilon_d-\omega_1}\left[\frac{2}{(\varepsilon_p-\varepsilon_q)(\varepsilon_p-E_d)} - \frac{1}{(\varepsilon_p-\varepsilon_d-\omega_1)(\varepsilon_q-E_d)}\right], \tag{7}$$

$$\tilde{P}_K^{(3)} = (N-1)\mu^4\mathcal{E}_1\mathcal{E}_2^2\sum_{\mathbf{pq}}\frac{(1-n_p)n_q}{\varepsilon_p-\varepsilon_d-\omega_1}\left[\frac{2}{(\varepsilon_p-\varepsilon_q)(\varepsilon_p-E_d)} - \frac{1}{(\varepsilon_p-\varepsilon_d-\omega_1)(\varepsilon_q-E_d)}\right], \tag{8}$$

where N is the impurity level degeneracy. Here we introduced the effective impurity level $E_d = \varepsilon_d + \omega_2$. The first term, $\tilde{P}_0^{(3)}$, is the usual third-order polarization for *spinless* ($N = 1$) electrons [9]. The second term, $\tilde{P}_K^{(3)}$, originates from the suppression, due to the Hubbard repulsion U, of the contributions from doubly occupied impurity states. As indicated by the prefactor $(N-1)$, it comes from the additional intermediate states that are absent in the spinless case [see Fig. 1(b)].

Consider the first term in Eq. (8). The restriction of the sum over $\mathbf{q}$ to states *below* the Fermi level results in a logarithmic divergence in the absorption coefficient, $\alpha \propto \mathrm{Im}\tilde{P}$, at the absorption threshold, $\omega_1 = -\varepsilon_d$:

$$\mathrm{Im}\tilde{P}_K^{(3)} = (N-1)p_0\theta(\omega_1+\varepsilon_d)\frac{2\Delta}{\pi\delta\omega}\ln\left|\frac{D}{\omega_1+\varepsilon_d}\right|, \tag{9}$$

where $p_0 = \pi\mathcal{E}_1\mu^2 g$, $\delta\omega = \omega_1 - \omega_2$ is the pump-probe detuning, and $\Delta = \pi g\mu^2\mathcal{E}_2^2$ is the energy width characterizing the pump intensity, and D and g are the bandwidth and the density of states (per spin) at the Fermi level, respectively. Recalling that the linear absorption is determined by $\mathrm{Im}\tilde{P}^{(1)} = p_0\theta(\omega_1+\varepsilon_d)$, we see that it differs from Eq. (9) by a factor $\frac{2\Delta}{\pi\delta\omega}\ln|\frac{D}{\omega_1+\varepsilon_d}|$ (setting for simplicity $N = 2$). In other words, $\mathrm{Im}\tilde{P}^{(1)}$ and $\mathrm{Im}\tilde{P}_K^{(3)}$ become comparable when

$$\omega_1 + \varepsilon_d \equiv \delta\omega + E_d \sim D \exp\left(-\frac{\pi\delta\omega}{2\Delta}\right). \qquad (10)$$

We see that the perturbative expansion of the nonlinear optical polarization in terms of the optical fields *breaks down* even for weak pump intensities (i.e., small Δ). The above condition of its validity depends critically on the detuning of the pump frequency from the Fermi level. For off-resonant pump, such that the effective impurity level lies below the Fermi level, $|E_d| = |\varepsilon_d| - \omega_2 \gg \Delta$, the relation (10) can be written as $\delta\omega + E_d \sim T_K$ with

$$T_K = De^{\pi E_d/2\Delta} = D \exp\left[-\frac{|\varepsilon_d| - \omega_2}{2g\mu^2\mathcal{E}_2^2}\right]. \qquad (11)$$

This new energy scale can be associated with the Kondo temperature—an energy scale known to emerge from a spin-flip scattering of a FS electron by a magnetic impurity [10]. Remarkably, in our case, the Kondo temperature can be *tuned* by varying the frequency and intensity of the pump. In fact, the logarithmic divergence in Eq. (9) is an indication of an *optically induced* Kondo effect.

Let us now turn to the second term in Eq. (8). In fact, it represents the lowest order in the expansion of the linear polarization with the impurity level shifted by $\delta\varepsilon = (N-1)\mu^2\mathcal{E}_2^2 \sum_{\mathbf{q}} \frac{n_q}{\varepsilon_q - E_d}$,

$$\tilde{P}^{(1)} = \mu^2\mathcal{E}_1 \sum_{\mathbf{p}} \frac{(1-n_p)}{\varepsilon_p - \varepsilon_d + \delta\varepsilon - \omega_1}. \qquad (12)$$

The origin of $\delta\varepsilon$ can be understood by observing that, for *monochromatic* pump, the coupling between the FS and the impurity can be described by a *time-independent* Anderson Hamiltonian H_A with effective impurity level $E_d = \varepsilon_d + \omega_2$ and hybridization parameter $V = \mu\mathcal{E}_2$. By virtue of this analogy, $\delta\varepsilon$ is the perturbative solution of the following equation for the self-energy part:

$$E_0 = \Sigma(E_0) \equiv (N-1)\mu^2\mathcal{E}_2^2 \sum_{\mathbf{q}} \frac{n_q}{\varepsilon_q - E_d + E_0}$$
$$\simeq (N-1)\frac{\Delta}{\pi} \ln\frac{E_d - E_0}{D}, \qquad (13)$$

which determines the renormalization of the effective impurity energy, E_d, to $\tilde{E}_d = E_d - E_0$ [10]. Indeed, to the first order in the optical field, Eq. (13) yields $E_0 = \delta\varepsilon$ after omitting E_0 in the right-hand side (rhs).

The logarithmic divergence (9) indicates that, near the absorption threshold, a nonperturbative treatment is necessary. Recall that the attractive interaction v_0 between a localized hole and FS electrons also leads to a logarithmically diverging correction (in the lowest order in v_0) even in the linear absorption: $\delta\tilde{P}^{(1)} \sim \tilde{P}^{(1)} g v_0 \ln[D/(\omega_1 + \varepsilon_d)]$. In the nonperturbative regime, $\delta\tilde{P}^{(1)} \sim \tilde{P}^{(1)}$, this correction evolves into the Fermi edge singularity [1]. The question is how the Kondo correction (9) will evolve in the nonperturbative regime. We first discuss our results qualitatively and defer the details to the end of the paper.

It can be seen from the expression (11) for T_K that there is a well-defined critical pump intensity, $\Delta_c \equiv \pi g \mu^2 \mathcal{E}_{2c}^2 = \frac{\pi}{2}(|\varepsilon_d| - \omega_2)$. The shape of the nonlinear absorption spectrum will depend sharply on the ratio between Δ and Δ_c. For strong pump, $\Delta > \Delta_c$, the Kondo correction (9) will develop into a broad peak with width Δ and height p_0. This is illustrated in Fig. 2(a).

Much more delicate is the case $\Delta \ll \Delta_c$, which is analogous to the Kondo limit. The Kondo scale T_K is then much smaller than Δ, which is the case for well-below-resonance pump excitation, $|\varepsilon_d| - \omega_2 \gg \Delta$. The impurity density of states in the Kondo limit is known [10] to have two peaks well separated in energy by $|E_d| = |\varepsilon_d| - \omega_2 \gg \Delta$ (E_d is the effective level position). As a result, in the presence of the pump, the system sustains excitations originating from the beats between these peaks. These excitations can assist the absorption of a probe photon. The corresponding condition for the probe frequency reads $|E_d| + \omega_1 \simeq |\varepsilon_d|$, or $\omega_1 \simeq \omega_2$. Thus, in the Kondo limit, the absorption spectrum exhibits a narrow peak below the linear absorption onset. This is illustrated in Fig. 2(b).

To calculate the shape of the below-threshold absorption peak, we adopt the large N variational wave-function method by following the approach of [11]. For monochromatic optical fields, the polarization (2) can be written as $\tilde{P} = -\mu^2\mathcal{E}_1[G^<(E_0 - \delta\omega) + G^>(E_0 + \delta\omega)]$, where $G^<(\varepsilon) = \langle\Omega|T^\dagger(\varepsilon - H_A)^{-1}T|\Omega\rangle$ [$G^>(\varepsilon)$ is similar but with $T \leftrightarrow T^\dagger$]. In the leading order in N^{-1}, $|\Omega\rangle$ is given by $|\Omega\rangle = A(|0\rangle + \sum_{\mathbf{q}} n_q a_q|\mathbf{q},1\rangle)$, where $|\mathbf{q},1\rangle = N^{-1/2}\sum_\sigma d_\sigma^\dagger c_{\mathbf{q}\sigma}|0\rangle$ ($|0\rangle$ represents the full FS). The coefficients A and a_k are found by minimizing H_A in

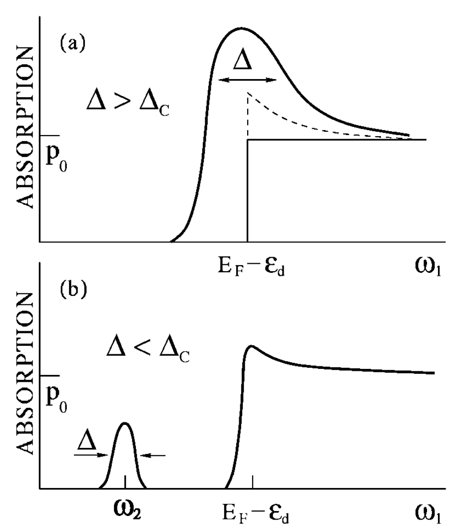

FIG. 2. Schematic plot of the nonlinear absorption spectra vs probe frequency. (a) Mixed-valence regime: pump-probe spectrum for strong pump intensity (thick line) compared with $\chi^{(3)}$ approximation (9) (dashed line) and the linear absorption spectrum (thin line). (b) Kondo limit: the pump-probe spectrum has a narrow peak below the linear absorption threshold.

this basis; one then obtains, e.g., $A^2 = 1 - n_d$, where $n_d = (1 + \pi\tilde{E}_d/N\Delta)^{-1}$ is the impurity occupation [10,11] ($N\Delta$ is finite in the large N limit). The relevant Green function is obtained as

$$G^{<}(\varepsilon) = \frac{\pi}{\Delta}\left[\Sigma(\varepsilon) + \frac{|\Sigma(\varepsilon)|^2}{\varepsilon - \Sigma(\varepsilon)}\right]. \tag{14}$$

Since $\Sigma(E_0) = E_0$ [see Eq. (13)], for $\varepsilon = E_0 - \delta\omega$ the second term has a pole at $\delta\omega = 0$ which gives rise to a resonance. The N^{-1} correction gives a finite resonance width Δ. Using that the residue at the pole is $[\partial\Sigma(E_0)/\partial E_0 - 1]^{-1} = n_d - 1$ [10], we finally obtain

$$\mathrm{Im}\tilde{P}_K = \frac{p_0 E_0^2(1 - n_d)^2}{\delta\omega^2 + \Delta^2} \sim \left(\frac{\pi E_d T_K}{N\Delta}\right)^2 \frac{p_0}{\delta\omega^2 + \Delta^2}. \tag{15}$$

For the last estimate, we used that, in the Kondo limit ($\Delta \ll \Delta_c$), $1 - n_d \simeq \pi T_K/N\Delta$ and $E_0 \simeq E_d$. Then the rhs of (15) describes the narrow below-threshold peak [see Fig. 2(b)]. In the Kondo limit, the factor $(1 - n_d)^2$ has the physical meaning of a product of populations of electrons in the narrow peak of the impurity spectral function (Kondo resonance) and "holes" in the wide peak (centered at ε_d below the Fermi level). Note, however, that the above calculation was not restricted to the Kondo limit. For $\Delta \gtrsim \Delta_c$ (mixed-valence regime), we have $1 - n_d \sim 1$ and $E_0 \sim N\Delta$. Then Eq. (15) reproduces the absorption peak in Fig. 2(a).

Note that, although we considered here, for simplicity, the limit of the singly occupied impurity level in the ground state, the Kondo absorption can take place even if the impurity is *doubly* occupied. Indeed, after the probe excites an impurity electron, the spin-flip scattering of FS electrons with the remaining impurity electron will lead to the Kondo resonance in the final state of the transition. In this case, however, the Kondo effect should show up in the fifth-order polarization.

A feasible system in which the proposed effect might be observed is, e.g., GaAs/AlGaAs superlattice delta doped with Si donors located in the barrier. The role of impurity in this system is played by a shallow acceptor, e.g., Be. Molecular-beam epitaxy growth technology allows one to vary the quantum well width and to place acceptors right in the middle of each quantum well [12]. In quantum wells, the valence band is only doubly degenerate with respect to the total angular momentum J. Thus, such a system emulates the large U limit considered here. The dipole matrix element for acceptor to conduction band transitions can be estimated as $\mu \sim \mu_0 a$, where μ_0 is the interband matrix element and a is the size of the acceptor wave function. For typical excitation intensities [2], the parameter Δ ranges on the meV scale resulting in $T_K \sim \Delta$ for the pump detuning of several meV.

In conclusion, let us discuss the effect of a finite duration of the pump pulse τ. Our result for $\chi^{(3)}$ remains unchanged if τ is longer than $\hbar/T_K$. If $\tau < \hbar/T_K$, then τ will serve as a cutoff of the logarithmic divergence in (9), and the Kondo correction will depend on the parameters of the pump $\mathcal{E}_2$ and τ as follows: $\mathrm{Im}\tilde{P}_K^{(3)} \propto \mathcal{E}_2^2 \ln(D\tau/\hbar)$. In the nonperturbative regime, our basic assumption was that, for monochromatic pump, the system maps onto the *ground* state of the Anderson Hamiltonian. Our results apply if the pump is turned on slowly on a time scale longer than $\hbar/T_K$. For a shorter pulse duration, the buildup of the optically induced Kondo effect will depend on the dephasing of FS excitations [8]. The role of interactions between FS and impurity electrons in the presence of hybridization was addressed in [13]. An avenue for future studies would be the interplay between the Kondo absorption and the Fermi edge singularity. Note finally that the effect of irradiation on the Kondo transport in quantum dots was investigated in [14,15].

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