Spin dynamics in nonlinear optical spectroscopy of Fermi sea systems

T. V. Shahbazyan
Department of Physics and Astronomy, Vanderbilt University, Nashville, TN 37235

I. E. Perakis
Department of Physics, University of Crete, P.O. Box 2208, 710 03, Heraklion, Crete, Greece

M. E. Raikh
Department of Physics, University of Utah, Salt Lake City, UT 84112

We discuss the role of many-body spin correlations in nonlinear optical response of a Fermi sea system with a deep impurity level. Due to the Hubbard repulsion between electrons at the impurity, the optical transitions between the impurity level and the Fermi sea states lead to an optically-induced Kondo effect. In particular, the third-order nonlinear optical susceptibility logarithmically diverges at the absorption threshold. The shape of the pump-probe spectrum is governed by the light-induced Kondo temperature, which can be tuned by varying the intensity and frequency of the pump optical field. In the Kondo limit, corresponding to off-resonant pump excitation, the nonlinear absorption spectrum exhibits a narrow peak below the linear absorption onset.

I. INTRODUCTION

The role of many-body correlations in the nonlinear optical spectroscopy of Fermi sea systems has attracted much interest during the last decade [1–3]. Due to the development of high quality ultrashort laser pulses, it has become possible to probe the elementary excitations of an interacting system on time scales shorter than the dephasing and relaxation times. In undoped semiconductors, it has been established that exciton-exciton interactions play a dominant role in the coherent regime [1]. Very recently, there has been a growing interest in studying the coherent dynamics of Fermi sea (FS) systems at low temperatures [4–9].

In this paper, we discuss a new many-body effect in the nonlinear optical response of a FS system with a deep impurity level. Specifically, we will focus on nonlinear absorption due to optical transitions between a localized impurity level and the continuum of FS states. In the linear absorption, two prominent many-body effects has long been known in such systems [10]. 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.

The nonlinear absorption comes from multiple transitions between impurity level and the Fermi sea. In particular, a number of different intermediate processes contribute to the third-order optical susceptibility \( \chi^{(3)} \). What is crucial for us here is that, in the system under study, some of the intermediate states involve the 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). Important is that, while on the impurity, the two electrons experience a Hubbard repulsion. In fact, such a repulsion gives leads to a logarithmic divergence in \( \chi^{(3)} \). The origin of such an anomaly is intimately related to the Kondo effect [12].

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.

We identify two distinct regimes which are characterized by the interplay between pump detuning and pump intensity. These regimes are somewhat analogous to the Kondo limit and mixed-valence regime in the “usual” Kondo systems. The shapes of nonlinear absorption spectra in these regimes are drastically different. The crossover between the two regimes is governed by a new energy scale – the light-induced Kondo temperature, which can be tuned by varying the intensity and frequency of the pump.

The paper is organized as follows. In Section II, we derive the Kondo-induced contribution to the third-order optical polarization that determines the absorption spectrum. In Section III, we obtain the absorption coefficient beyond \( \chi^{(3)} \) using variational large \( N \) method. Section IV concludes the paper.
Polarization then takes the form $P$ and can be excluded from the expansion of the polarization (3) with respect to a singly-occupied $U$ considered sufficiently large values of $i\partial$ equation $\mu$ direction and central frequency, respectively, and $\epsilon$ transition channels are possible from states below the FS to the two spin-degenerate states of empty impurity, but only one channel from the singly-occupied impurity to states above the FS.

II. KONDO ANOMALY IN $\chi^{(3)}$

We start with the Hamiltonian of the system: $H_{\text{tot}} = H + H_1(t) + H_2(t)$, where

$$H = \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \epsilon_d \sum_\sigma d_{\sigma}^\dagger d_\sigma + \frac{U}{2} \sum_{\sigma \neq \sigma'} \hat{n}_\sigma \hat{n}_{\sigma'},$$

is the Hamiltonian in the absence of optical fields; here $c_{\mathbf{k}\sigma}^\dagger$ and $d_\sigma^\dagger$ are conduction and localized electron creation operators, respectively, $(\hat{n}_\sigma = d_{\sigma}^\dagger d_\sigma)$; $\epsilon_{\mathbf{k}}$ and $\epsilon_d$ 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

$$H_i(t) = -M_i(t) \hat{T}^\dagger + \text{h.c.}, \quad \hat{T}^\dagger = \sum_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}^\dagger d_\sigma, \quad M_i(t) = e^{i\mathbf{k} \cdot \mathbf{r} - i\omega_i t} \mu \mathcal{E}_i(t),$$

where $\hat{T}^\dagger$ is the optical transition operator. Here $\mathcal{E}_i(t)$, $\mathbf{k}_i$, and $\omega_i$ are the pump/probe electric field amplitude, direction and central frequency, respectively, and $\mu$ is the dipole matrix element ($i = 1, 2$ denotes the probe and pump, respectively). 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 [11]:

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

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 [3] with respect to $H_2$. The third-order pump-probe polarization then takes the form $P^{(3)}(t) = e^{i\mathbf{k}_1 \cdot \mathbf{r} - i\omega_1 t} \hat{P}^{(3)}$ with

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

where

FIG. 1. Intermediate processes contributing to $\chi^{(3)}$. (a) Intermediate state with doubly-occupied impurity: excitation of a FS electron to the second impurity state followed by the excitation of the two impurity electrons to the FS. (b) Large $U$ limit: Two transition channels are possible from states below the FS to the two spin-degenerate states of empty impurity, but only one channel from the singly-occupied impurity to states above the FS.
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

$$F(t, t', t_1, t_2) = \langle \Omega_0|\hat{T}e^{-iH(t-t_1)}\hat{T}\rangle e^{-iH(t'-t_1)}\hat{T}\rangle|\Omega_0\rangle = \sum_{pqk'k\lambda\sigma'} A_{pqk'k}^{\lambda\sigma'\sigma}\epsilon^{-(i\epsilon_p-i\epsilon_d)(t-t')-i(i\epsilon_k-i\epsilon_d)(t-t_1)-i(i\epsilon_k-i\epsilon_d)(t_1-t_2)},$$

(6)

with $n_\sigma = \langle \Omega_0|d_{\sigma}^\dagger d_{\sigma}|\Omega_0\rangle$ and $n_k = \langle \Omega_0|c_{k\sigma}^\dagger c_{\sigma}|\Omega_0\rangle$ (impurity occupation number is $n_d = \sum_\sigma n_\sigma = 1$ here). Eqs. (3) and (7) are valid in the large $U$ limit corresponding to a singly-occupied impurity. For monochromatic optical fields, $\mathcal{E}_i(t) = \mathcal{E}_1$, the time integrals can be explicitly evaluated,

$$Q_1(t, t') = \mathcal{E}_2^2 \sum_{pq} e^{-i(\epsilon_p-\epsilon_d)(t-t')}(1-n_p) \left[ \frac{2Nn_q}{(\epsilon_p-\epsilon_d)(\epsilon_p-E_d)} - \frac{i(t-t')(1-n_q)}{(\epsilon_q-E_d)} \right],$$

$$Q_2(t, t') = \mathcal{E}_2^2 \sum_{pq} e^{i(\epsilon_p-\epsilon_d)(t-t')}(1-n_p) \left[ \frac{i(t-t')Nn_q}{(\epsilon_q-E_d)} + \frac{2(1-n_q)}{(\epsilon_p-\epsilon_q)(\epsilon_p-E_d)} + \frac{(1-n_q)e^{i(\epsilon_p-E_d)(t-t')}}{(\epsilon_q-E_d)(\epsilon_p-E_d)} \right],$$

$$Q_3(t, t') = -\mathcal{E}_2^2 \sum_{pq}(1-n_p) \left[ \frac{e^{i(\epsilon_p-\epsilon_q-\omega_2)(t-t')Nn_q + e^{-i\omega_2(t-t')(1-n_q)}}}{(\epsilon_q-E_d)(\epsilon_p-E_d)} \right],$$

(8)

yielding $\tilde{\mathcal{P}}(3) = \tilde{\mathcal{P}}_0(3) + \tilde{\mathcal{P}}_K(3)$ with

$$\tilde{\mathcal{P}}_0(3) = \mu^4 \mathcal{E}_1^2 \sum_{pq} \left( \frac{1-n_p}{\epsilon_p-\epsilon_d-\omega_1} \right) \left[ \frac{2}{(\epsilon_p-\epsilon_q)(\epsilon_p-E_d)} - \frac{1}{(\epsilon_p-\epsilon_d-\omega_1)(\epsilon_q-E_d)} \right],$$

$$\tilde{\mathcal{P}}_K(3) = (N-1)\mu^4 \mathcal{E}_1^2 \sum_{pq} \left( \frac{1-n_p}{\epsilon_p-\epsilon_d-\omega_1} \right) \left[ \frac{2}{(\epsilon_p-\epsilon_q)(\epsilon_p-E_d)} - \frac{1}{(\epsilon_p-\epsilon_d-\omega_1)(\epsilon_q-E_d)} \right],$$

(9)

(10)

where $N$ is the impurity level degeneracy. Here we introduced the effective impurity level $E_d = \epsilon_d + \omega_2$. The first term, $\tilde{\mathcal{P}}_0(3)$, is the usual third-order polarization for spinless ($N = 1$) electrons [11]. The second term, $\tilde{\mathcal{P}}_K(3)$, 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. (11). The restriction of the sum over $q$ to states below the Fermi level results in a logarithmic divergence in the absorption coefficient, $\alpha \propto \text{Im} \tilde{\mathcal{P}}$, at the absorption threshold, $\omega_1 = -\epsilon_d$:

$$\text{Im} \tilde{\mathcal{P}}_K(3) = (N-1)p_0\theta(\omega_1 + \epsilon_d) \frac{2\Delta}{\pi\delta\omega} \ln \left| \frac{D}{\omega_1 + \epsilon_d} \right|,$$

(11)

where $p_0 = \pi \mathcal{E}_1^2 g$, $\delta\omega = \omega_1 - \omega_2$ is the pump-probe detuning, and $\Delta = \pi g\mu^2 \mathcal{E}_2^2$ is the energy width characterizing the pump intensity; $D$ 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 $\text{Im} \tilde{\mathcal{P}}(1) = p_0\theta(\omega_1 + \epsilon_d)$, we see that it differs from Eq. (11) by a factor $\frac{2\Delta}{\pi\delta\omega} \ln \left| \frac{D}{\omega_1 + \epsilon_d} \right|$ (setting for simplicity $N = 2$). In other words, $\text{Im} \tilde{\mathcal{P}}(1)$ and $\text{Im} \tilde{\mathcal{P}}_K(3)$ become comparable when
\[ \omega_1 + \varepsilon_d \equiv \delta \omega + E_d \sim D \exp \left( -\frac{\pi \delta \omega}{2 \Delta} \right). \]  

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 \( \Delta \)). 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 (12) can be written as \( \delta \omega + E_d \sim T_K \) where

\[ T_K = D e^{\pi \varepsilon_d/2 \Delta} = D \exp \left( -\frac{|\varepsilon_d| - \omega_2}{2g \mu^2 \varepsilon_0^2} \right). \]  

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 [13]. 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. (11) is an indication of an optically-induced Kondo effect.

Let us now turn to the second term in Eq. (10). In fact, it represents the lowest order in the expansion of the linear polarization with impurity level shifted by \( \delta \varepsilon \equiv (N-1)\mu^2 \varepsilon_0^2 \sum_q n_q \varepsilon_{q-d} \).

\[ \tilde{P}^{(1)} = \mu^2 \varepsilon_0 \sum_p \frac{(1-n_p)}{\varepsilon_p - \varepsilon_d + \delta \varepsilon - \omega}, \]  

(14)

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 \( V = \mu \varepsilon_2 \). By virtue of this analogy, \( \delta \varepsilon \) itself is the perturbative solution of the following equation for the self-energy part [3]

\[ E_0 = \Sigma(E_0) \equiv (N-1)\mu^2 \varepsilon_0^2 \sum_q \frac{n_q}{\varepsilon_q - E_d + E_0} \simeq (N-1)\frac{\Delta}{\pi} \ln \frac{E_d - E_0}{D}, \]  

(15)

which determines the renormalization of the effective impurity energy, \( E_d \), to \( \tilde{E}_d = E_d - E_0 \) [3]. Indeed, to the first order in the optical field, Eq. (15) yields \( E_0 = \delta \varepsilon \) after omitting \( E_0 \) in the rhs.

The logarithmic divergence (14) indicates that near the absorption threshold, a nonperturbative treatment is necessary. Recall that, in the lowest order in \( \nu_0 \), the attractive interaction \( \nu_0 \) between a localized hole and FS electrons also leads to a logarithmically diverging correction even in the linear absorption: \( \delta \tilde{P}^{(1)} \sim \tilde{P}^{(1)} \nu_0 \ln[D/(\omega_1 + \varepsilon_d)] \). In the nonperturbative regime, \( \delta \tilde{P}^{(1)} \sim \tilde{P}^{(1)} \), this correction evolves into the Fermi edge singularity [10]. The question is how the Kondo correction (11) will evolve in the nonperturbative regime. This question is addressed in the next section.

### III. General Shape of Absorption Spectrum

We start by discussing qualitatively the results and defer the details to the end of the section.

It can be seen from the expression (3) for \( T_K \) that there is a well-defined critical pump intensity,

\[ \Delta_c \equiv \pi g \mu^2 \varepsilon_0^2 = \frac{\pi}{2} \left( |\varepsilon_d| - \omega_2 \right). \]  

(16)

Note that \( |\varepsilon_d| - \omega_2 \) is the pump detuning from the Fermi level. The shape of the nonlinear absorption spectrum depends sharply on the ratio between \( \Delta \) and \( \Delta_c \). The strong pump case, \( \Delta > \Delta_c \), is analogous to mixed-valence regime. In this regime, the Kondo correction (11) develops into a broad peak with width \( \Delta \) and height \( p_0 \). This is illustrated in Fig. 2(a).

Much more delicate is the case \( \Delta < \Delta_c \), which is analogous to the Kondo limit. The Kondo scale \( T_K \) is then much smaller than \( \Delta \), 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 [3] 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, in fact, 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).
FIG. 2. Schematic plot of the Kondo-absorption spectra vs probe frequency. (a) Mixed-valence regime: spectrum for strong pump intensity (thick line) compared with the perturbative result (dashed line) and the linear absorption spectrum (thin line). (b) Kondo limit: the nonlinear absorption spectrum exhibits a narrow peak below the linear absorption threshold.

To calculate the shape of the below-threshold absorption peak, we adopt the large $N$ variational wave-function method by following the approach of Ref. [14]. For monochromatic optical fields, the polarization (3) can be written as

$$\tilde{P} = -\mu^2 \mathcal{E}_1 \left[ G^<(E_0 - \delta \omega) + G^>(E_0 + \delta \omega) \right], \quad (17)$$

where

$$G^<(\epsilon) = \langle \Omega | T^\dagger (\epsilon - H_A)^{-1} T | \Omega \rangle, \quad (18)$$

and $G^>(\epsilon)$ is similar but with $T \leftrightarrow T^\dagger$. In the leading order in $N^{-1}$, the ground state $|\Omega\rangle$ is given by

$$|\Omega\rangle = A \left( |0\rangle + \sum_q n_q a_q |q, 1\rangle \right), \quad (19)$$

where $|q, 1\rangle = N^{-1/2} \sum_{\sigma} \hat{d}_{q}^\sigma c_{q \sigma} |0\rangle$ ($|0\rangle$ stands for the full FS). The coefficients $A$ and $a_q$ are found by minimizing $H_A$ in this basis,

$$A^2 = 1 - n_d, \quad a_q = \frac{\sqrt{N} \mu \mathcal{E}_2}{E_d - \epsilon_q - E_0}, \quad (20)$$

where

$$n_d = \left( 1 + \frac{\pi E_d}{N \Delta} \right)^{-1} \quad (21)$$

is the impurity occupation [14,15] ($N \Delta$ is finite in the large $N$ limit). Using that $\hat{T} |\Omega\rangle = \sqrt{N} \sum_q |q, 1\rangle$, the Green function $G^<(\epsilon)$ takes the form

$$G^<(\epsilon) = NA^2 \sum_{pq} n_p n_q \langle p, 1 | \frac{1}{\epsilon - H_A} | q, 1 \rangle, \quad (22)$$
where the matrix element in the rhs is obtained, to the leading order in \( N^{-1} \), by inverting \( H_A \) in the above basis set,

\[
\langle p', 1 | \frac{1}{\epsilon - H_A} | q, 1 \rangle = \frac{\delta_{pq}}{\epsilon - \epsilon_{q} - E_d} + \frac{1}{\epsilon - \Sigma(\epsilon)} (\epsilon + \epsilon_{q} - E_d)(\epsilon + \epsilon_{p} - E_d),
\]

with \( \Sigma(\epsilon) \) given by Eq. (13). We then obtain

\[
G^\ast (E_0 - \delta \omega) = \frac{\pi}{\Delta} \left[ \Sigma(E_0 - \delta \omega) + \frac{|\Sigma(E_0 - \delta \omega)|^2}{E_0 - \delta \omega - \Sigma(E_0 - \delta \omega)} \right].
\]

Since \( \Sigma(E_0) = E_0 \) [see Eq. (15)], 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 \( \Delta \). The first term gives nonresonant contribution to absorption for \( \delta \omega \) 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 \( \epsilon_d \) below the Fermi level). In this case, it can be shown that contribution from \( G^\ast (E_0 + \delta \omega) \) is suppressed by factor \( N^{-1} \) and is nonresonant.

Near the resonance, using that \( [\partial \Sigma(E_0)/\partial E_0 - 1]^{-1} = n_d - 1 \) \( \sim \), we obtain

\[
\text{Im} \tilde{P}_K = \frac{E_0^2(1 - n_d)^2}{\delta \omega^2 + \Delta^2}.
\]

For \( \Delta \ll \Delta_c \), corresponding to the Kondo limit, we have \( 1 - n_d \approx \pi T_K/N\Delta \) and \( E_0 \approx E_d \), so that

\[
\text{Im} \tilde{P}_K \sim p_0 \left( \frac{\pi E_d T_K}{N\Delta} \right)^2 \frac{1}{\delta \omega^2 + \Delta^2}.
\]

In this case, \( (26) \) describes the narrow below-threshold peak [see Fig. 2(b)]. 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 \( \epsilon_d \) below the Fermi level). In the opposite case, \( \Delta \gtrsim \Delta_c \), corresponding to mixed-valence regime, we have \( 1 - n_d \approx 1 \) and \( E_0 \approx N\Delta \). Then the polarization

\[
\text{Im} \tilde{P}_K = \frac{N^2\Delta^2}{\delta \omega^2 + \Delta^2}
\]

(27) describes the absorption peak in Fig. 2(a).

**IV. CONCLUSION**

Note that, although we considered here, for simplicity, the limit of 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. MBE growth technology allows one to vary the quantum well width and to place acceptors right in the middle of each quantum well \([13]\). 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 \( \mu_0 \) is the interband matrix element and \( a \) is the size of the acceptor wave function. For typical excitation intensities \([2]\), the parameter \( \Delta \) ranges on the meV scale resulting in \( T_K \sim \Delta \) for the pump detuning of several meV.

Finally, let us discuss the effect of a finite duration of the pump pulse, \( \tau \). Our result for \( \chi^{(3)} \) remains unchanged if \( \tau \) is longer than \( \hbar/T_K \). If \( \tau < \hbar/T_K \), then \( \tau \) will serve as a cutoff of the logarithmic divergence in \( [14] \), and the Kondo correction will depend on the parameters of the pump \( \epsilon_2 \) and \( \tau \) as follows: \( \text{Im} \tilde{P}_K^{(3)} \propto \epsilon_2^2 \ln(D\tau/\hbar) \). In the non-perturbative 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 shorter pulse duration, the build up of the optically-induced Kondo effect will be determined by the dynamics of FS excitations.

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