Effect of Kondo resonance on optical third harmonic generation

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Abstract

We use the method of dynamical mean field theory, to study the effect of Kondo resonance on optical third harmonic generation (THG) spectra of strongly correlated systems across the metal-insulator transition. We find that THG signals are proportional to the quasiparticle weight $z$ of the Kondo peak, and are precursors of Mott-Hubbard gap formation.

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Keywords Kondo resonance; Third harmonic generation; Metal-insulator transition

1 Introduction

One of the earliest and most important triumphs of the Dynamical mean field theory (DMFT) \cite{1} was to elucidate the nature of metal-to-insulator transition (MIT) driven by strong electron correlation. It was demonstrated that this transition is accompanied by a Kondo resonance at the Fermi surface \cite{2}, width of which is proportional to the quasiparticle weight $z$, and approaches to zero as the Mott insulating side ($U \rightarrow U_c^-$) is approached from the metallic side. Therefore in nonlinear optical spectra, we expect some signals that vanish in the limit $U \rightarrow U_c^-$, as a result of quasiparticle weight $z \rightarrow 0$.

Equivalently one can employ the pressure to tune $U_c$ and hence switch from Mott insulating side to a regime in which such signals arise due to Kondo peak. This is a unique feature of correlated insulators, that has no analogue in band-insulators. In case of band insulators, to generate quasiparticle states in the mid-gap, one may need to dope them, while in Mott insulators such mid-gap states can be created by applying pressure to a Mott insulator which is in the onset of MIT. Possible examples would be a class of materials known as Kondo insulators \cite{3}. (A typical example of this family is YbB$_{12}$ \cite{4}).

2 Method of calculation

One of the applications of nonlinear optical materials is the third harmonic generation (THG) \cite{5}, that requires materials capable of providing strong enough nonlinear optical signals. To study this process in correlated electronic systems, we have developed the theory of nonlinear optical response in the context of DMFT \cite{6} which is also a suitable theoretical tool to study the effect of Kondo resonance in nonlinear spectra. In $d \rightarrow \infty$ limit where DMFT becomes exact,
vertex corrections to current operators identically vanish \[8\] so that the THG susceptibility within DMFT approximation becomes \[6\]

\[
\chi_{\text{THG}}(\nu) = \tilde{\tau} \frac{4}{6\pi \nu^4} \int d\omega d\varepsilon D(\varepsilon) \frac{1}{\xi_0 - \varepsilon} \frac{1}{\xi_1 - \varepsilon} \frac{1}{\xi_2 - \varepsilon} \frac{1}{\xi_3 - \varepsilon},
\]

where \(\xi_j = \omega + j\nu - \Sigma_R(\omega + j\nu) + i|\Sigma_I(\omega + j\nu)|\) for \(j = 0, 1, 2, 3\), and \(D(\varepsilon) = 2\sqrt{1 - \varepsilon^2}/\pi\) is the semicircular DOS corresponding to the normalized hopping \(\tilde{\tau} = 1/2\).

In the above formula, (i) the integration over \(\varepsilon\) corresponds to summation over intermediate states in conventional expressions \[5\] which are usually used for systems with discrete energy levels, and (ii) the matrix element effects are encoded in \(\Sigma(\omega)\), real and imaginary part of which have been denoted by \(\Sigma_R\) and \(\Sigma_I\), respectively. It is very crucial to note that we have used absolute value of the imaginary part of the self-energy. This is indeed a necessary step to transform from time-ordered four-current, to fully retarded one \[7\].

We obtain self-energy \(\Sigma\) by solving the iterated perturbation theory equations of DMFT for the semi-circular density of states \[9\]. In DMFT approximation, this quantity is purely local.

The critical value of Hubbard \(U\) corresponding to semicircular Bethe lattice is \(U_c \sim 3.35\), above which we are in Mott insulating phase (Fig. 1). As can be seen in Fig. 1, the sharp Kondo resonance state at Fermi surface in the onset of transition to Mott insulating phase, is characteristic of correlation driven MIT.

3 Results

Fig. 2 shows the imaginary part of the THG susceptibility \(\chi_{\text{THG}}(\nu)\) of eq. (1) as a function of incident photon energy \(\nu\) for various values of Hubbard \(U\) close to MIT. Dotted line corresponds to \(U = 3.5\) which is in the Mott insulating phase. The Mott-Hubbard gap is manifested by a region in which \(\text{Im}\chi_{\text{THG}}\) is identically zero. The onset of three-photon absorption as demonstrated earlier \[6\] corresponds to the frequency where \(\text{Im}\chi_{\text{THG}}\) takes up, and is 1/3 of the Mott-Hubbard gap \((E_g)\).

Three-photon resonances in this formulation correspond to the peak \(A\) in \(\text{Im}\chi_{\text{THG}}\) around \(\nu \sim 0.8\) (dotted line), while two-photon features appear as a dip \(B\) around \(\nu \sim 1.7\) \[6\]. As can be seen in Fig. 4 except for slight shift in the location of the dip \(B\), the two-photon features are not much affected by moving to lower values of \(U\) in \(U \lesssim U_c\) region. In contrast, the three-photon features is entirely distorted as we cross the \(U_c\) and enter the Kondo regime of \(U \lesssim U_c\).

Below the critical value \(U_c \sim 3.35\) the system is in metallic side, with a sharp Kondo peak present at the Fermi surface \[2\]. For \(U = 3.3\) (dashed line) just below the critical value, there appear peaky structures below \(\nu \sim 1\). Among these structures, weak features denoted by \(A'\) and \(B'\) in Fig. 2 start to appear in the THG ‘gap’ region \((0 < \nu < E_g/3)\). These features seem to correspond to three-photon and two-photon resonances between the lower Hubbard band and the Kondo resonance state (schematically depicted in Fig. 1), as they vanish.
Figure 1: Density of states within DMFT at the onset of transition to insulating state (dashed line) and in the Mott insulating state (dotted line). A and A’ represent schematically the three-photon resonance processes in $\chi^{\text{THG}}$ of Fig. 2. Such three-photon resonances appear as peaks A and A’ in Fig. 2 as explained in Ref. 6. Similarly B and B’ in this figure schematically represent two-photon resonances denoted by B and B’ in Fig. 2.

in $z \rightarrow 0$ limit. Moving further away from MIT, quasiparticle weight $z$ grows and hence A’ and B’ features also start to grow proportional to $z$. To see this proportionality, note that $\chi^{\text{THG}}$ in eq. 11 is dominated by $D(\varepsilon)$ effect, and hence in the limit $z \rightarrow 0$ one can replace the Kondo peak with a sharp peak of strength $z$. Then decomposing to partial fractions, each with a single $(\xi_j - \varepsilon)$ denominator in eq. 11, one can see that $j$-photon features for $j = 1, 2, 3$ will be proportional to $z$.

Implication of this observation for Kondo insulators is that, an external pressures on the order of a fraction of GPa can lead to instability in the functioning of optical devices built on them by filling in the gap and hence reducing the contrast of the output signal.

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Figure 2: Imaginary part of $\chi^{\text{THG}}$ across the Mott metal-to-insulator transition. Critical value of $U$ is $\sim 3.35$. Solid line corresponds to $U = 3.2$, dashed line corresponds to $U = 3.3$, and dotted line corresponds to $U = 3.5$, which is in the Mott insulating side of MIT transition. The onset of three-photon absorption for $U = 3.5$ is where imaginary part of $\chi^{\text{THG}}$ starts to become non-zero (around $\nu \sim 0.5$).

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