Terahertz excited plasmon-magnon interaction and magnetoplasmon-enhanced energy transfer within the framework of generalized spin Hamiltonian

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Abstract. Research into ultrafast terahertz technology attracts a significant interest Nowadays in the fields of plasmonics and magnonics, since a recent progress in both fields unveils new fundamental physics and opens up new opportunities to engineering high-speed spintronic devices. However insufficient exchange of knowledge between these research fields still remains. The goal of this paper is to bridge such a gap by presenting a new theoretical approach to study magnetoplasmon-enhanced energy transfer between supported magnetic nanoparticles in the framework of generalized spin Hamiltonian (GSH) under ultrafast THz-wave excitation.

1. Introduction

Ultrafast spin dynamics induced by transport of spin polarized carriers has attracted considerable interest over last decade [1, 2]. It is motivated by the fundamental interest in magnetic excitations and applications like spintronics and data storage[3, 4]. To achieve an understanding of the underlying elementary processes at microscopic scale, that typically occur on femtosecond time scales, a time dependent approach has been developed that probes the spin dynamics induced by plasmon-magnon excitations[5]. Surface magnetoplasmons (SMP) in the THz range are technologically important because they provide a possibility to develop subwavelength-size devices and offer an opportunity to investigate nanoscale systems at THz frequencies time-domain spectroscopy. In conjunction with pump-probe measurements it offers a powerful tool to examine energy transfer and ultrafast spin dynamics in nanosystems. The THz spectral range provides a high degree of sensitivity for research of a spin structure, which at very strong exchange complex generally dominate the charge response[6]. Further, with an ultrafast optical excitation, this approach permits one to probe non-equilibrium systems with picosecond to sub-picosecond time resolution. These unique capabilities of the optical-pump/THz-probe
spectroscopy give the possibilities for the investigation of energy transfer phenomena that have remained inaccessible with conventional techniques.

Magnons can be interpreted as collective excitations associated to the electron spin structure and plasmons can be directly linked to high-energy oscillations of the electron density in the cluster. In this context, a magnon is a spin wave with zero phase, and it can be understood as a quantum of a spin wave. Both quasiparticles (magnon and plasmon) and their collective excitations are of great importance for better understanding of the electronic origin of magnetic phenomena and, therefore, the study of their properties is a key prerequisite regarding the applications in charge-based electronics and spintronics[7].

In this work we describe the process of Förster transfer (FT) between nanoparticles in the presence of a metal nanocrystal subsystem [8]. We focus here on an optical processes, which take place on a Ni/Cu(100) surface in the vicinity of $\text{Gd}_3\text{N@C}_{80}$ endohedral metallofullerenes nanoparticles (EMFNPs). Since, it was shown, that such nanoparticles can strongly modify the optical process [9]. EMFNPs on a Ni/Cu(001) surface strongly confine carriers and do not permit efficient tunnel coupling. However, instead of direct tunnel coupling, EMFNPs permit FT of magnetic generated magnons. The FT mechanism comes from the inter-EMFNP Coulomb interaction and does not require the tunnel coupling between EMFNPs. The enhancement of Förster transfer can occur due to the effect of plasmon-assisted amplification of electric fields inside the nanoscale assembly [10]. Here, we show how the theory of Förster resonant energy transfer (FRET) may be generalized through inclusion the spin structure of a d-,f-shell nanoparticles. In particular, here we demonstrate that the photoluminescence spectra should be carefully analyzed since the plasmon-enhanced Förster effect can appear together with strong magnon energy dissipation.

2. Theoretical approach
To study magnetoplasmon-enhanced energy transfer between interacted EMFNPs on a Ni/Cu(001) surface, we used numerical scheme constructed from two parts. The first part is represented by the re-expanded model [11] used to describes the electrostatic interaction between two charged (polarized) metallofullerene $\text{Gd}_3\text{N}$ nanoparticle spheres surrounded by a homogeneous dielectric medium $\epsilon_d$. Let us to consider these nanoparticle spheres charged by $q_B$ and $q_A$ with relative dielectric functions $\epsilon_{\text{EMFNP}}(\omega)$ and $\epsilon_{\text{EMFNP}}(\omega)$, and radii $R_B$ and $R_A$, respectively. The center-center distance between spheres is labeled by $R$ and the spheres are surrounded by a medium with dielectric constant $\epsilon_d$, as shown in Fig. 1.

Since we study the energy transfer between a donor (d) Ni ion and an acceptor (a) Cu ion on copper substrate in the vicinity of two EMFNPs, the donor and acceptor ions will be represented by point electric dipoles $\mathbf{p}_d$ and $\mathbf{p}_a$.

When two charged or polarized spheres are located close together, an infinite number of multipoles are induced in the spheres. These multipoles lead to the difficulties in the precise evaluation of the enhancement factor between these two spheres [12]. Therefore we used the re-expansion of a given two Legendre expansion for the electrostatic field in a Taylor series around
the two centers of the nanoparticle spheres [11]. If a dipole \( \mathbf{p} \) is located at the position \( P \), it will induce polarization charges on both spheres. The potential at the position of the dipole charge is then given by:

\[
\Phi_{\text{out}} = \sum_{l=0}^{\infty} \left[ c_l^B \left( \frac{R_B}{r_B} \right)^{l+1} P_l(\cos \Theta_B) + c_l^A \left( \frac{R_A}{r_A} \right)^{l+1} P_l(\cos \Theta_A) \right] + \sum_{l=0}^{\infty} \frac{1}{2} \left[ \delta c_l^B \left( \frac{R_B}{r_B} \right)^{l+1} P_l(\cos \Theta_B) + \delta c_l^A \left( \frac{R_A}{r_A} \right)^{l+1} P_l(\cos \Theta_A) \right],
\]

where \( P_l \) denote associated Legendre functions and \( c_l^A/B \) are coefficients in decompositions. The first sum is the contribution from the sphere–sphere interaction, which can be determined by solving next equations:

\[
(\mathbf{H}_{A/B} - \mathbf{I}) \mathbf{c}^{A/B} = \mathbf{w}^{A/B},
\]

where \( \mathbf{I} \) is the unit matrix; \( \mathbf{c}^A \) and \( \mathbf{c}^B \) are two column vectors with elements \( c_l^A \) and \( c_l^B \), respectively. The matrix and vector elements are:

\[
h^{A/B}_{lm} = f(\epsilon_{\text{EMFNP}A/B}, l) \left( \frac{R_A}{R} \right)^{l+m+1} \left( \frac{R_B}{R} \right)^{j_{\text{max}}} \sum_{j=1}^{j_{\text{max}}} f(\epsilon_{\text{EMFNP}B/A}, j) \left( \frac{R_B}{R} \right)^{2j},
\]

\[
w^A_{l} = f(\epsilon_{\text{EMFNP}A/B}, l) \frac{1}{4\pi \epsilon_d R} \left( \frac{R_A}{R} \right)^l \left( \frac{R_B}{R} \right)^{j_{\text{max}}} \sum_{j=1}^{j_{\text{max}}} f(\epsilon_{\text{EMFNP}B/A}, j) \left( \frac{R_B}{R} \right)^{2j},
\]

\[
f(\epsilon, j) = \frac{j(\epsilon / \epsilon_d - 1)}{j(\epsilon / \epsilon_d + 1) + 1}.
\]

In the same manner we obtained two matrix equations:

\[
(\delta \mathbf{H}_{A/B} - \mathbf{I}) \delta \mathbf{c}^{A/B} = \delta \mathbf{w}^{A/B},
\]

where

\[
\delta \mathbf{H}_{A/B} = \mathbf{H}_{A/B}
\]

\[
\delta w^B_{l} = f(\epsilon_{\text{EMFNP}B}, l) \frac{Q(r)}{4\pi \epsilon_d r} \left[ \frac{1}{r} \left( \frac{R_B}{r} \right)^l - \frac{R_A}{R^2 - R r} \sum_{j=1}^{j_{\text{max}}} f(\epsilon_{\text{EMFNP}A}, j) \frac{(j+l)!}{j!!} \left( \frac{R_A^2}{R^2 - R r} \right)^j \right] - \frac{R_A}{R^2 - R r} \sum_{j=1}^{j_{\text{max}}} f(\epsilon_{\text{EMFNP}A}, j) \frac{(j+l)!}{j!!} \left( \frac{R_A^2}{R^2 - R r} \right)^j
\]

\[
\delta w^A_{l} = f(\epsilon_{\text{EMFNP}A}, l) \frac{Q(r)}{4\pi \epsilon_d (R - r)} \left[ \frac{1}{R - r} \left( \frac{R_A}{R - r} \right)^l - \frac{R_B}{R r} \sum_{j=1}^{j_{\text{max}}} f(\epsilon_{\text{EMFNP}B}, j) \frac{(j+l)!}{j!!} \left( \frac{R_B^2}{R r} \right)^j \right] - \frac{R_B}{R r} \sum_{j=1}^{j_{\text{max}}} f(\epsilon_{\text{EMFNP}B}, j) \frac{(j+l)!}{j!!} \left( \frac{R_B^2}{R r} \right)^j
\]
The second part of our numerical scheme shows how FRET can be generalized with including the spin structure of a d-,f-shell nanoparticles. The influence of the electric and magnetic component of the electromagnetic (EM) field can be described in the framework of GSH:

\[ \hat{H} = \hat{H}_{\text{ion}}^{(o)} + \hat{H}_{\text{spin}}^{(o)} + \hat{H}'(t) \]  

(9)

where

\[ \hat{H}_{\text{ion}}^{(o)} = -\nabla^2 + V_{\text{ion}} \hat{e}_{\text{eff}} + \hat{H}_{\text{ex}} + \hat{H}_{\text{an}} + \hat{H}_{\text{ZEE}}, \hat{H}'(t) = E_0 e^{-i\omega t} - \gamma S \hat{H}_{\text{eff}}^{\text{LLG}} e^{-i\omega t}. \]  

(10)

Here \( V_{\text{ion}} \hat{e}_{\text{eff}} \) represents the effective single-electron potential of ion associated with \( Q(r) = -rV_{\text{ion}}(r)/2 \). Hamiltonians \( \hat{H}_{\text{ex}}, \hat{H}_{\text{an}}, \) and \( \hat{H}_{\text{ZEE}} \) denote the Heisenberg isotropic exchange interaction, the anisotropic exchange interaction due to the axial single-ion anisotropy, and the interaction between the spin system and the external magnetic field, respectively. The last term of equation describes consequently the electric and magnetic contributions of EM field, where \( \gamma \) is the gyromagnetic ratio coming from the relation between spin \( \gamma = \frac{e\hbar}{2m} \).

The solution [13, 14]:

\[ \frac{\partial \langle \hat{S} \rangle}{\partial t} = \frac{1}{1 + \lambda^2} \langle \hat{S} \rangle \times \hat{H}_{\text{eff}}^{\text{LLG}} - \frac{\lambda}{1 + \lambda^2} \langle \hat{S} \rangle \times (\langle \hat{S} \rangle \times \hat{H}_{\text{eff}}^{\text{LLG}}), \]  

(11)

where \( \lambda \) is damping constant. The eigenstates \( |SM\rangle \) of GSH are given by the linear combinations of the basis states \( |S^{(\mu)}M^{(\mu)}\rangle \), where \( M = -S, ..., S, \mu = 1, 2, ..., N - 1 \), and the coefficients \( c_{\mu\nu} \) can be evaluated once the spin Hamiltonian of the system has been diagonalized. With the algebra of the spin operators we can obtain the expectation values for \( \hat{H}_{\text{spin}}^{(o)} \):

\[ \langle \hat{S}_x \rangle = \frac{1}{2} \sum_{\mu=1}^{N} c_{\mu\mu}^2 A_{\mu}, \langle \hat{S}_y \rangle = -\frac{i}{2} \sum_{\mu=1}^{N} c_{\mu\mu}^2 B_{\mu}, \langle \hat{S}_z \rangle = \sum_{\mu=1}^{N} c_{\mu\mu}^2 M^{(\mu)}, \]  

(12)

where

\[ A_{\mu} = \sqrt{S^{(\mu)}(S^{(\mu)} + 1) - M^{(\mu)}(M^{(\mu)} + 1) + 1} \sqrt{S^{(\mu)}(S^{(\mu)} + 1) - M^{(\mu)}(M^{(\mu)} + 1) - 1} \]  

and

\[ B_{\mu} = \sqrt{S^{(\mu)}(S^{(\mu)} + 1) - M^{(\mu)}(M^{(\mu)} + 1) - 1} \sqrt{S^{(\mu)}(S^{(\mu)} + 1) - M^{(\mu)}(M^{(\mu)} + 1) + 1}. \]  

Now we turn to the discussion about the photoresponse of EMFNPs to presence of an external EM field in terms of the frequency-dependent polarizability \( \alpha(\omega) \). For this purpose we analyze at first the dispersion properties of the surface magnetoplasmons (SMP) in the EMFNPs structure under an applied external magnetic field. The dielectric response in the THz frequency region originates from the intraband motion of conduction electron and spins. Therefore we can use Drude model to describe the THz electron response and the Kramers-Heisenberg dispersion formula for \( \alpha(\omega) \). In this case, a dielectric function \( \epsilon(\omega) \) for magnetoplasmon system is:

\[ \epsilon_{(x,y,z)}(\omega) = \epsilon_{(x,y,z)}^{\text{Drude}}(\omega) + \alpha_{(x,y,z)}^{\text{spin}}(\omega) \]  

(13)
In the framework of Drude model, the components of the dielectric tensor in presence of magnetic field are the following:

\[
\hat{\epsilon}_{xx}^{\text{Drude}} = \epsilon_b \left[ 1 - \frac{\omega_p^2}{(\omega^2 + i\omega\Gamma)} \right], \quad \hat{\epsilon}_{xy}^{\text{Drude}} = \frac{i\omega \epsilon_b \omega_c}{(\omega^2 + i\omega\Gamma)^2 - \omega^2 \omega_c^2}, \quad \hat{\epsilon}_{xx}^{\text{Drude}} = \epsilon_b \left[ 1 - \frac{\omega_p^2}{\omega^2 + i\omega\Gamma} \right],
\]

where \(\epsilon_b\) is the background dielectric constant, \(\omega_p\) is the effective plasma frequency, which lies in the terahertz regime, \(\omega_c\) is the cyclotron frequency, and \(\Gamma\) is the damping constant.

The Kramers-Heisenberg dispersion formula can be applied to calculate \(\alpha_{\text{spin}}(\omega)[10]\). So far as, the Hamiltonian \(\hat{H}_{\text{spin}}^{(i)}\) is induced by transitions between different spin substates of the magnetic molecule, let us denote:

\[
M_{i_f}^{(z)} = \sum_{\nu=1}^{N} c_{\nu} c_{\nu f} A_{\nu}; M_{i_f}^{(y)} = \sum_{\nu=1}^{N} c_{\nu} c_{\nu f} B_{\nu}; M_{i_f}^{(x)} = \sum_{\nu=1}^{N} c_{\nu} c_{\nu f} M_\nu
\]

where \(i\) is initial spin energy state \(|SM\rangle_i\) and \(f\) is final spin energy \(|SM\rangle_f\) of GSH in the spin structure. Then

\[
\alpha_{\text{spin}}^{(x)}(\omega) = 2\gamma^2 \sum_{i=1}^{N_i} \sum_{f=1}^{N_f} |H_{i_f}^{(LLG)}(x) M_{i_f}^{(x)}|^2 \frac{\omega_f - \omega_i}{(\omega_f - \omega_i)^2 - (\omega - i\Gamma)^2},
\]

\[
\alpha_{\text{spin}}^{(x)}(\omega) = 2\gamma^2 \sum_{i=1}^{N_i} \sum_{f=1}^{N_f} H_{i_f}^{(LLG)}(x) H_{i_f}^{(LLG)}(x) M_{i_f}^{(x)} M_{i_f}^{(x)} \frac{\omega_f - \omega_i}{(\omega_f - \omega_i)^2 - (\omega - i\Gamma)^2}.
\]

Thus, for the dielectric response in the THz frequency region we obtain:

\[
\epsilon_{xx} = \hat{\epsilon}_{xx}^{\text{Drude}}(\omega) + (4\pi \alpha_{xx}^{\text{spin}}(\omega) + 1), \quad \epsilon_{xz} = \hat{\epsilon}_{xz}^{\text{Drude}}(\omega) + (4\pi \alpha_{xz}^{\text{spin}}(\omega) + 1)
\]

In this EMFNP structure the Gd3N cluster is gyroelectric anisotropic with permittivity tensor:

\[
\hat{\epsilon}_{EMFNP} = \begin{pmatrix}
\epsilon_{xx} & 0 & \epsilon_{xz} \\
0 & \epsilon_{yy} & 0 \\
-\epsilon_{xz} & 0 & \epsilon_{zz}
\end{pmatrix}
\]

Further, we describe energy transfer between Ni (a donor point electric dipole \(p_d\) at locations \(r_d\)) and Cu (an acceptor point electric dipole \(p_a\) at locations \(r_a\)) ions in the vicinity of EMFNPs on a Ni/Cu/001 surface. For this purpose, we propose here a theory of the plasmon-magnon enhancement of the many body phenomena resulting for the SMP-dressed Coulomb interaction. The interaction energy between the Ni and Cu ions (dipoles \(p_a\) and \(p_d\), respectively) can be obtained from the expression:

\[
U = -\frac{1}{2} \left| p_a \cdot E_a + p_d \cdot E_d \right|,
\]

where \(E_a\) is the electric field at the position of the acceptor ion and \(E_d\) is the corresponding field at the donor ion. These fields are obtained using

\[
E = -\nabla \Phi_{\text{out}}
\]

and omitting the terms \(\Phi_a\), when computing \(E_a\), and \(\Phi_d\), when computing \(E_d\). Thus we may write:

\[
\Delta U(\omega) = -\frac{1}{2} \left| p_a \cdot E_d(r_a, \omega) + p_d \cdot E_a(r_d, \omega) \right|,
\]
where $E_j(r_i, \omega)$ is the field of the jth particle at the point $r_i$.

Finally, we examine our numerical scheme in direct calculations of FRET between interacted EMFNPs including SMP, as illustrated schematically in Fig. 1. For selected interparticle distance and radii we observed an energy transfer between considered particles mediated predominantly via SMP (see Fig 2). We revealed a significant enhancement of magnetoplasmon-enhanced energy transfer if the particles increase their size (see Fig. 3). Thus, the obtained results clearly show that even a small variation in distance can effect an energy transfer between magnetic nanoparticles, that could be important to develop the ultrafast spintronic devices.

**Figure 2.** (Color online) The energy transfer between EMFNP1 ($R_B = 0.22\text{nm}$) and EMFNP2 ($R_A = 0.22\text{nm}$) on a Ni/Cu(001) surface.

**Figure 3.** (Color online) The energy transfer between EMFNP1($R_B = 0.24\text{nm}$) and EMFNP2($R_A = 0.24\text{nm}$) on a Ni/Cu(001) surface.

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