Polariton mediated Raman scattering in microcavities: a Green’s function approach

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Abstract

We present calculations of the intensity of polariton-mediated inelastic light scattering in semiconductor microcavities within a Green’s function framework. In addition to reproducing the strong coupling of light and matter, this method also enables the inclusion of damping mechanisms in a consistent way. Our results show excellent agreement with recent Raman scattering experiments.

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I. INTRODUCTION

The strong coupling between light and exciton modes in semiconductor microcavities (MC’s) has attracted a great deal of interest in the last decade as a novel means to study and control the interaction between radiation and matter [1, 2, 3, 4, 5, 6, 7, 8, 9, 10]. The strong coupling of photon and exciton modes in MC’s leads to the formation of the polariton modes which are quasiparticles formed by a quantum superposition of light and matter. Polariton physics is clearly displayed in resonant Raman scattering (RRS) experiments in which the wave length of the incoming radiation is tuned in such a way that the energy of the outgoing radiation coincides with that of a polariton mode, after emitting a longitudinal optical (LO) phonon, for the same conserved two-dimensional (2D) in-plane wave vector \( k_\parallel \) [4, 5, 6, 7, 8, 9, 10]. Former simplified theories, which consider polaritons as the eigenstates of a two-level model, can only qualitatively explain these experiments, and point clearly into the relevance of including dephasing and damping effects in these descriptions [6, 7, 8, 9].

In this work we describe the effects of including the coupling with the electron-hole continuum, and the damping effects in RRS as mentioned in reference [11]. We consider a system where only two exciton-polariton branches are observed [8], which means that only the 1s exciton plays an important role. A survey of the formalism is presented in the Section II. Section III contains the results and discussion, and finally some conclusions are presented in Section IV.

II. MODEL

We use the following Hamiltonian to model the system [11]

\[
H = E_f f^\dagger f + E_e e^\dagger e + (V e^\dagger f + H.c.) + \sum_p \epsilon_p r_p^\dagger r_p + \sum_p (V_p r_p^\dagger f + H.c.)
+ \sum_q \epsilon_q d_q^\dagger d_q + \sum_q (V_q d_q^\dagger e + H.c.) + \sum_k \epsilon_k c_k^\dagger c_k + \sum_k (V_k c_k^\dagger f + H.c.)
\]

(1)

where \( f^\dagger \) and \( e^\dagger \) are creation operators of the MC photon and exciton respectively. Momentum \( k_\parallel \), polarization and other quantum numbers have been omitted to simplify the notation. The first three terms describe the strong coupling between the MC photon and the exciton [6, 7, 8, 9]. The fourth and the fifth terms describe respectively a continuum of radiative modes, with creation operators \( r_p^\dagger \), and its coupling with the MC photon. The following two
terms have a similar effect for the exciton mode, and $d_q^\dagger$ describe a combined excitation due to scattering with other quasiparticles (e.g. acoustic phonons). Finally, the last two terms describe the electron-hole continuum (with $c_k^\dagger$ the creation operator of a single excitation with relative momentum $k = k_{\text{electron}} - k_{\text{hole}}$) and its coupling with the MC photon. This Hamiltonian can be diagonalized in the form $H = \sum_\nu E_\nu p_\nu^\dagger p_\nu$, where the boson operators $p_\nu^\dagger$ correspond to linear combinations of all creation operators entering Eq. (1). Denoting the latter for brevity as $b_j^\dagger$, then $p_\nu^\dagger = \sum_j A_{\nu j} b_j^\dagger$. Instead of diagonalizing the Hamiltonian we will work with retarded Green’s functions $G_{jl}(\omega) = \langle \langle b_j; b_l^\dagger \rangle \rangle_\omega$. These Green’s functions can be calculated with the equation of motion method, $\omega\langle \langle b_j; b_l^\dagger \rangle \rangle_\omega = \delta_{jl} + \langle \langle [b_j, H]; b_l^\dagger \rangle \rangle_\omega$.

To describe the Raman intensity we use previous models [7, 8], in which $I \propto T_i T_s W$, where $T_i$ is the probability of conversion of an incident photon $|f_i\rangle$ into a cavity eigenmode $|\nu_i\rangle$, $T_s$ has an analogous meaning for the scattered eigenmode $|\nu_s\rangle$ and the outgoing photon $|f_s\rangle$, and $H'$ is the interaction between electrons and the LO phonons. $W$ is the transition probability per unit of time, which we calculate with the Fermi’s Golden rule. Taking $T_s = |A_{\nu_s f}|^2$ (the weight of photon in the scattered eigenmode) and approximating $\langle \nu_i | H' | \nu_s \rangle \simeq A_{\nu_s e}$ (the excitonic part of the scattered eigenmode), we obtain $I = |A_{\nu_s e}|^2 |A_{\nu_s f}|^2 \rho(\omega)$ where $\rho(\omega) = \sum_j \rho_{jj}(\omega)$ is the density of final states, which can be calculated using the relation $\rho_{jj}(\omega) = -\frac{1}{\pi} \text{Im} G_{jj}(\omega)$, and where we have neglected the dependence of $T_i$ on frequency [11]. The quantities $A_{\nu ij}$ can straightforwardly be calculated as $|A_{\nu ij}|^2 = \rho_{jj}(\omega)/\rho(\omega)$.

When $\omega$ is chosen in such a way that the resonance condition for the outgoing polariton is fulfilled, we obtain [11]

$$I = \frac{\rho_{ff}(\omega) \rho_{ee}(\omega)}{\rho_{ff}(\omega) + \rho_{ee}(\omega)}. \quad (2)$$

The Green’s functions calculated from the equation of motion are

$$G_{ff}(\omega) = \frac{1}{w - E_f + i\delta_f - \frac{V^2}{w-E_e+i\delta_e} + S_f'}, \quad G_{ee}(\omega) = \frac{1}{w - E_e + i\delta_e - \frac{V^2}{w-E_f+i\delta_f} }, \quad (3)$$

where we have approximated the coupling with continuous modes $r_p^\dagger$ and $d_q^\dagger$ as purely imaginary contributions $-i\delta_f$ and $-i\delta_e$ respectively. The electron-hole continuum states enter through the sum $S_f'(\omega) = \sum_k \frac{|V_k|^2}{w+i\delta_f-\epsilon_k}$, and start contributing at the energy of the gap. This corresponds to vertical transitions in which the light promotes a valence electron with 2D wave vector $k$ to the conduction band with the same wave vector. Using the effective-mass approximation we obtain [11], $S_f' = R(\omega) - i(\omega - E_{XC})K\Theta(\omega - E_{XC})$, where $E_{XC}$ is the
bottom of the electron-hole continuum (the energy of the semiconductor gap) and $K$ is a dimensionless parameter that controls the magnitude of the interaction. The real part $R(\omega)$ can be absorbed in a renormalization of the photon energy and is unimportant in what follows. The imaginary part is a correction to the photon width $\delta_f$ for energies above the bottom of the continuum.

III. RESULTS AND DISCUSSION

Applying the theory outlined in the previous section, we calculated the Raman intensity for the case of a MC displaying two polariton branches, i.e. where only the 1s exciton mode interacts strongly with the cavity-photon mode, and a Rabi splitting of $2V \sim 19\,meV$. The parameters are chosen to be the same as those used in Ref. [11]. Our results, and the comparison with the experimentally measured RRS intensities are shown in Fig. 1. Figure 1a

![FIG. 1](image_url)

**FIG. 1:** (a) Calculated Raman profiles as function of the Stokes shifted energy for selected fixed positive detuning ($E_f - E_e > 0$), showing the LP and UP resonances. Dashed lines indicate the dispersion of the polariton branches (see text for details). (b) Raman intensity at exact resonance with the UP branch as a function of detuning. Solid squares: experimental results of Ref. [8]. Solid lines: theory (Eq. 2). Dashed line: result for a 2x2 matrix, neglecting damping effects.

shows the calculated Raman intensity profiles (as function of the Stokes shifted energy) for
some selected and fixed positive photon-exciton detuning \((E_f - E_e > 0)\). The calculated profiles show resonances at the energies corresponding to the lower (LP) and upper (UP) polariton branches, indicated by the dotted lines. The maximum of the resonance with the UP (indicated with circles in Fig. 1a), is plotted with a thick solid line in Fig. 1b as function of detuning. The experimental RRS data from Ref. [8] taken at exact resonance with the cavities UP is also displayed (solid squares). The agreement between experiment and theory is very good indeed. We can see that when the upper polariton (UP) mode energy increases above \(E_{XC}\) entering the electron-hole continuum (thick lines in Fig. 1a), the intensity of the Raman process falls abruptly. This can be understood from Eq. (3): the imaginary part in the denominator increases producing a decrease in values of \(\rho_{ff}(\omega)\), corresponding to processes in which light has enough energy to promote valence band electrons into the conduction band. The continuous creation and annihilation of electron-hole excitations produce a decrease of MC photon lifetime.

For completeness, we also performed analogous calculations for lower values of \(K\) (thin solid lines in Fig. 1b). In the limiting case of \(K \rightarrow 0\) we reobtain the expected symmetric curve (dotted line) corresponding to the case where no interaction with the electron-hole continuum is considered and damping effects are neglected.

IV. SUMMARY

We have presented a Green’s function approach to describe polariton mediated Raman scattering in experiments. This method allows a consistent introduction of damping mechanisms in polariton lifetime, being the most significant the interaction of MC photon with the electron-hole continuum. We are able to reproduce the main features of the experiment and the observed decrease in the Raman efficiency when the UP polariton branch enters the continuum of electron-hole excitations.
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