Nonlinear Optical Effects of a Coupled Semiconductor Quantum Dot - Metal Nanoparticle System

Andreas F. Terzis[1], Spyridon G. Kosionis[1], John Boviatsis[2] and Emmanuel Paspalakis[3]

[1] *Department of Physics, University of Patras, Patras 265 04, Greece*

[2] *Technological and Educational Institute of Western Greece, Megalou Alexandrou 1, Patras 263 34, Greece*

[3] *Materials Science Department, University of Patras, Patras 265 04, Greece*

SCOPE OF THIS WORK: The study of the nonlinear optical properties of complex nanosystems that involve the interaction of excitons from semiconductor quantum dots and surface plasmons from metallic nanostructures has attracted significant attention recently both theoretically and experimentally [1,2]. A structure that has attracted particular attention is a hybrid nanocrystal complex composed of a semiconductor quantum dot and a spherical metal nanoparticle. The optical response of this structure interacting with a weak probe field of varying frequency has been recently theoretically studied [3], and phenomena such as optical transparency and gain without inversion were identified in the linear susceptibility of the semiconductor quantum dot. In addition, it is found that the linear susceptibility of the metal nanoparticle is strongly influenced by the presence of the quantum dot. Here, we extend this study and present results for the third order, $\chi^{(3)}$, and fifth order, $\chi^{(5)}$, susceptibilities, using the nonlinear density matrix equations [1,3-5], including the contribution of both the semiconductor quantum dot and the metal nanoparticle. We then investigate the dependence of the $\chi^{(3)}$ and $\chi^{(5)}$ susceptibilities on the interparticle distance as well as on the material parameters of the quantum dot.

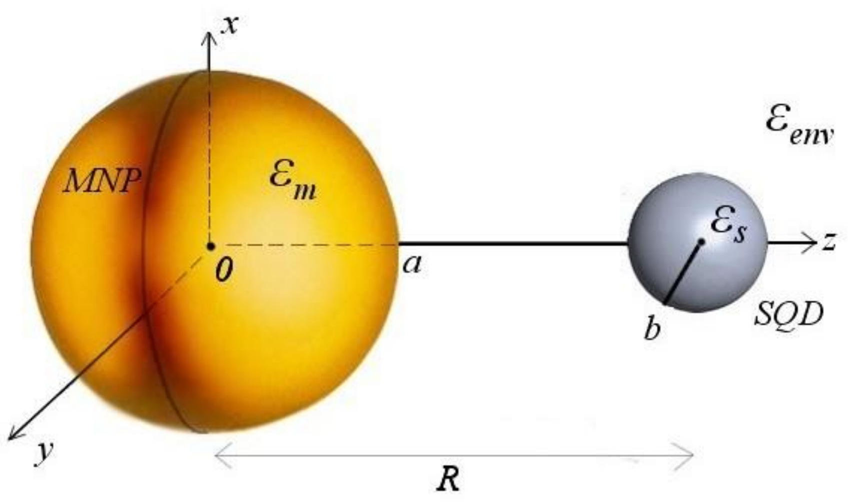

Figure 1: A coupled system consisting of a semiconductor quantum dot (SQD) and a spherical metal nanoparticle (MNP). The system interacts with a linearly polarized probe field. The field is taken polarized parallel to the interparticle axis.

CALCULATION METHODOLOGY: For the calculation we use the density matrix methodology (see, e.g., refs. [1,3-5]) for the present system. The interaction between excitons and surface plasmons is taken into account in the calculations [1-5]. We solve the density matrix equations in steady state analytically, using the method presented in Ref. [6], in order to properly account for the nonlinear dependence of the population difference and the polarization of the quantum dot on the electric field amplitude. Then, we use these results for presenting analytical formulae for the $\chi^{(3)}$ and $\chi^{(5)}$ susceptibilities for the quantum dot contribution, the metal nanoparticle contribution and the coupled quantum dot – metal nanoparticle system (total system).

MAIN RESULTS: Typical results of the $\chi^{(3)}$ and $\chi^{(5)}$ susceptibilities of the quantum dot, the metal nanoparticle and the coupled system for several interparticle distances, for a colloidal quantum dot (CdSe-based quantum dot) and gold nanoparticle are shown in Figs. 2-4. For the parameters used, see Ref. [3]. We see that the nonlinear optical susceptibilities are suppressed as the interparticle distance decreases. The opposite behavior is found for epitaxial quantum dots.

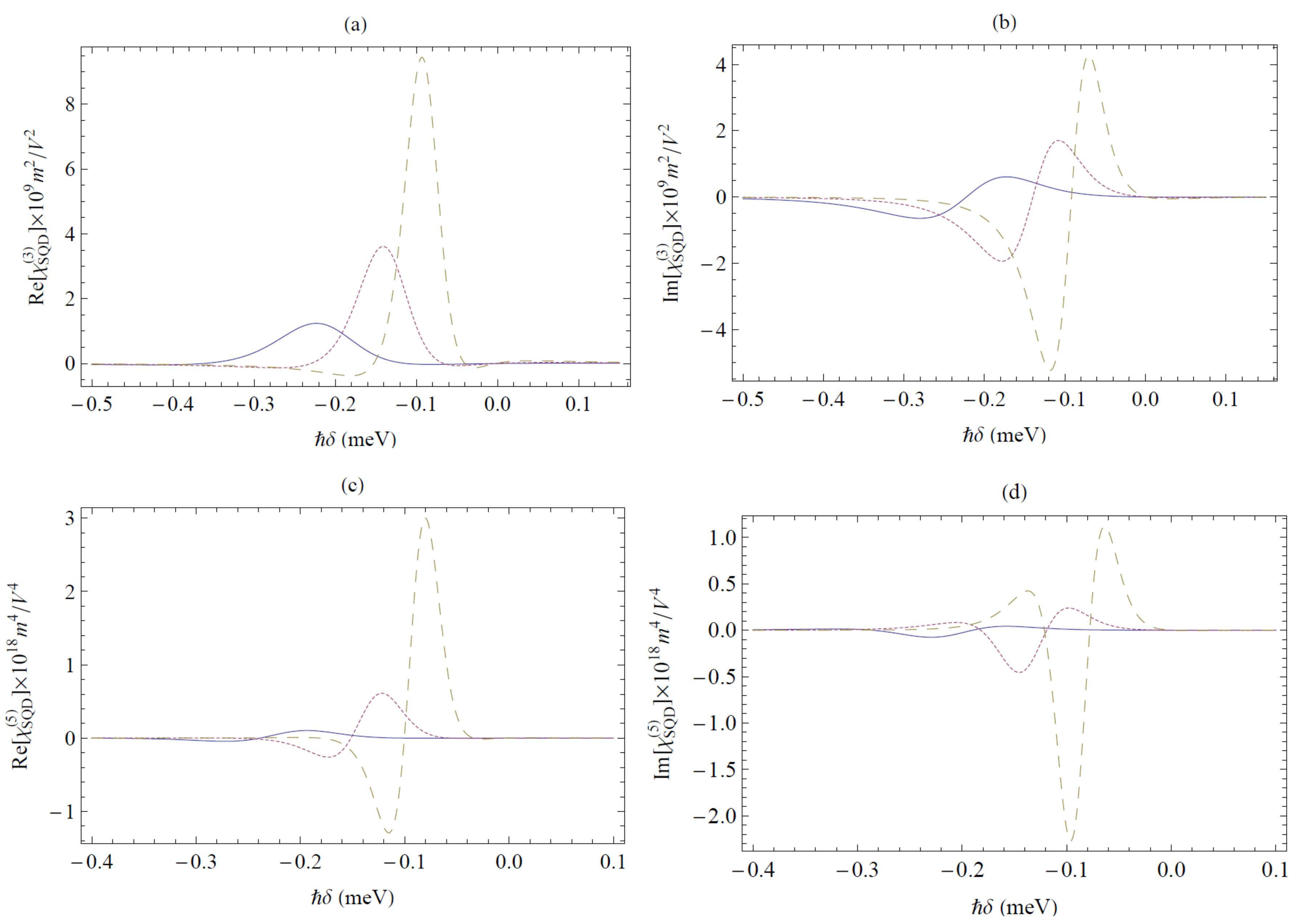

Figure 2: The real [(a) and (c)] and imaginary [(b) and (d)] parts of the $\chi^{(3)}$ [(a) and (b)] and $\chi^{(5)}$ [(c) and (d)] susceptibilities of the quantum dot contribution for a colloidal quantum dot for different values of the interparticle distance: R = 11.8 nm (solid curve), 12.4 nm (dotted curve) and 13 nm (dashed curve).

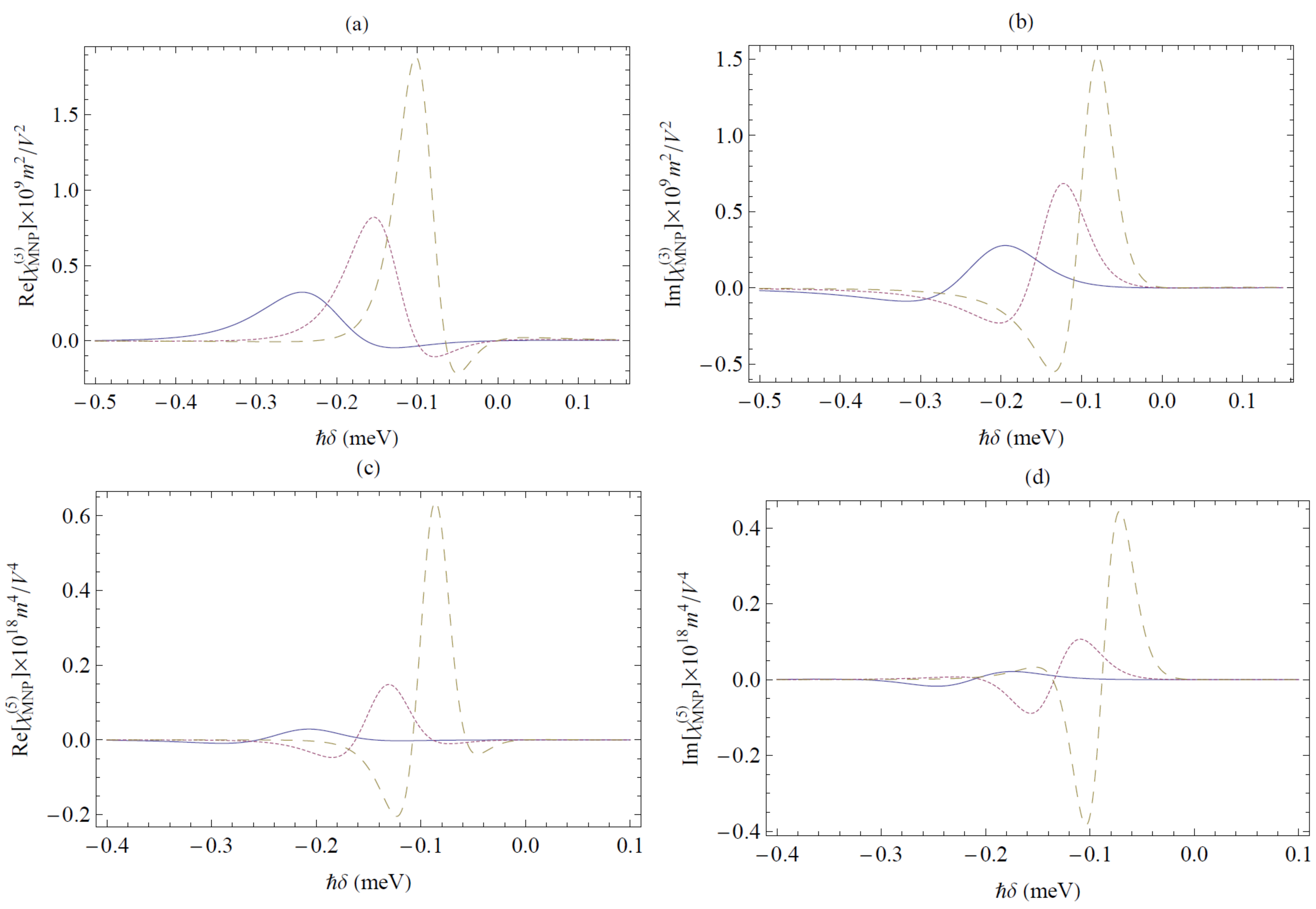

Figure 3: The same as in Fig. 2 but for the metal nanoparticle contribution.

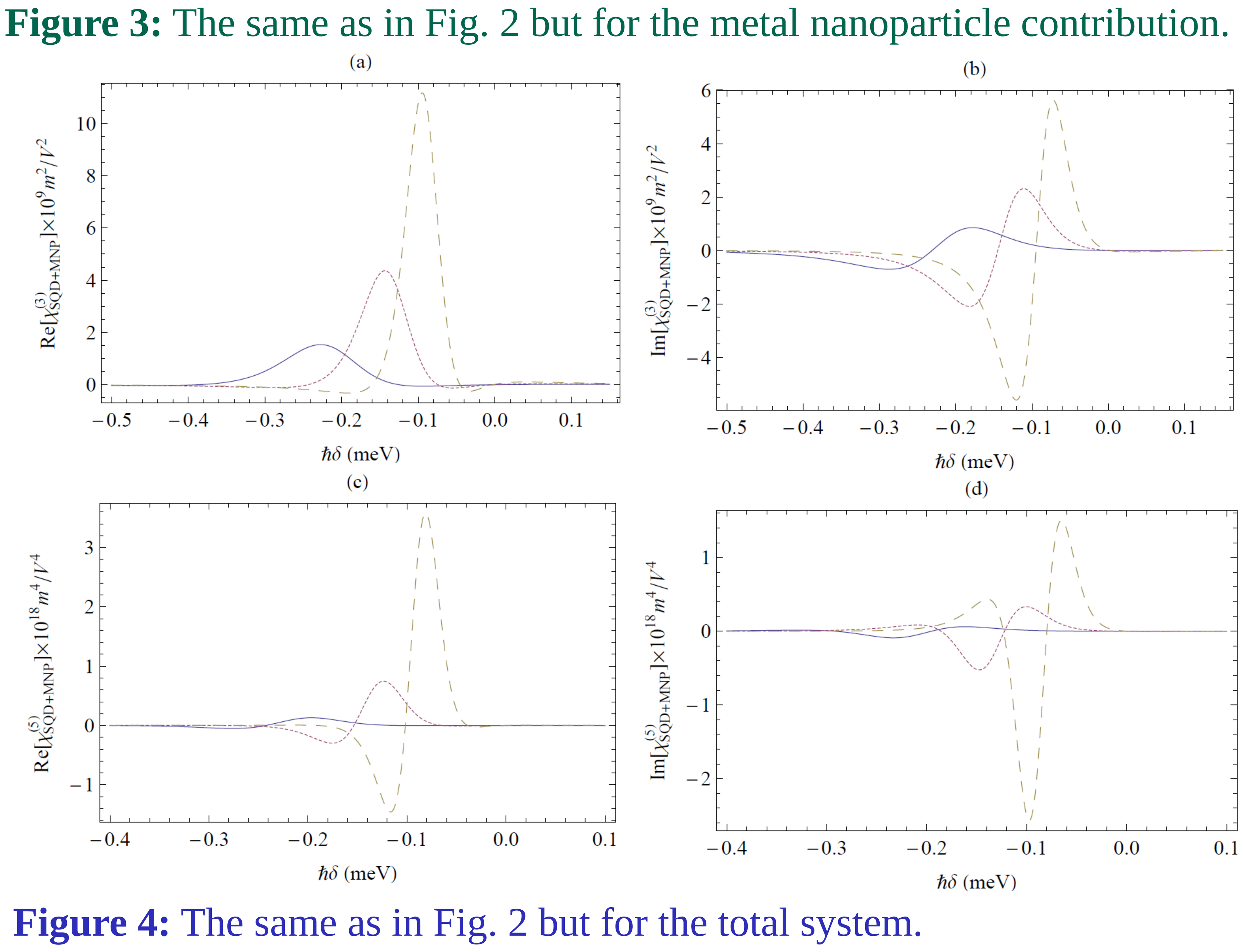

Figure 4: The same as in Fig. 2 but for the total system.

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