The fact that the low energy peak in the simulated Ag absorption spectra is more intense than the high energy peak is in line with the resonance strengths obtained from Eq. (1) for a spherical Ag NP in SubPc. It is however in contrast to the experimentally observed absorption differences between SubPc thin-films with and without Ag NPs, which could be due to damping of the low energy LSPR by the inhomogeneous size and shape distribution of the Ag NPs. Qualitatively, however, these results are well in agreement with the experimentally observed results and clearly confirm the presence of absorption enhancement in SubPc at wavelengths above the high energy LSPR as well as the presence of two plasmon resonances in the Ag NPs surrounded by SubPc. Furthermore, as observed experimentally, the low energy resonance peak is not present in the case of the uncovered truncated Ag spheroids [dashed curve in Fig. 3(b)]. Finally, the x and y components of the electric field surrounding the truncated Ag spheroids covered by 10 nm of SubPc shown in Fig. 3(c) confirm that both LSPRs are indeed dipolar in nature.

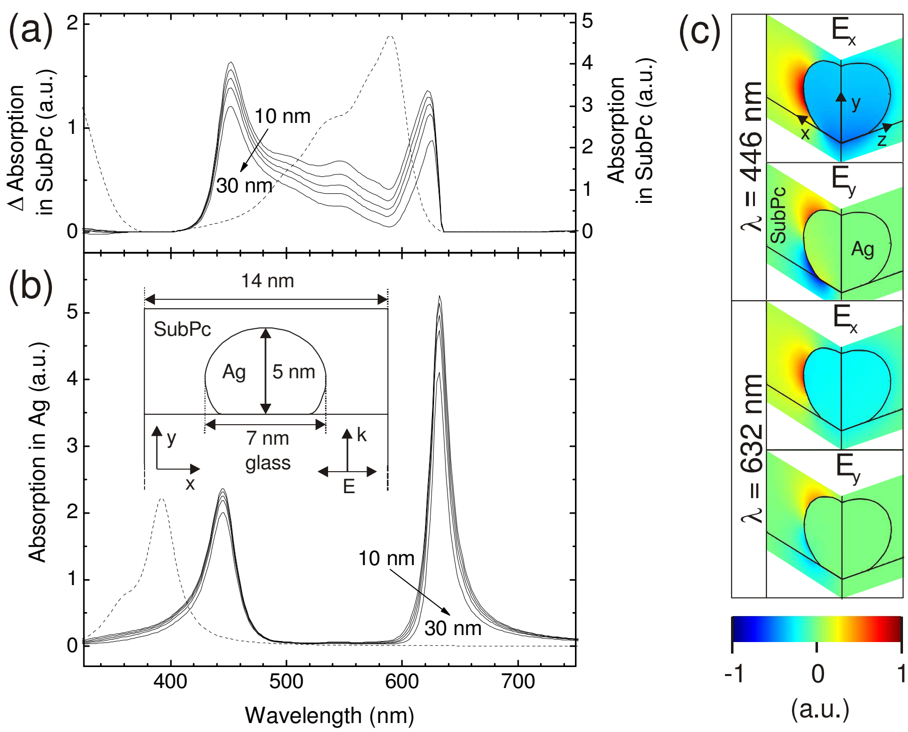

Fig. 3. Results of three-dimensional numerical simulations performed using a geometry with a cross-section as shown in the inset in (b). Periodic boundaries in both lateral dimensions result in an infinite two-dimensional array of truncated Ag spheroids with center-to-center spacing of 14 nm. The light wave is incident through the glass substrate with the propagation vector (**k**) and electric field vector **E** as indicated by arrows. (a) Difference between the absorption in the SubPc layers with thicknesses of 10, 14, 18, 22, and 30 nm covering the truncated Ag spheroids and the absorption in pure SubPc layers (excluding the volume corresponding to the truncated Ag spheroids) with the same thicknesses (solid curves). The absorption of a pure SubPc layer with a thickness of 10 nm is shown as a dashed curve. (b) Absorption in the uncovered truncated Ag spheroids (dashed curve) and in the truncated Ag spheroids covered by SubPc layers with the same thicknesses as in (a) (solid curves). (c) Surface plots of the x and y components of the electric field at λ = 446 and 632 nm for a SubPc thickness of 10 nm.

For similar metal NP-organic dye systems, the formation of hybridized exciton-plasmon polariton states caused by strong coupling has been reported, as evidenced by splitting of the absorption bands [22,29]. Indeed, both experimental results and simulations suggest the appearance of a weak shoulder at λ ~550 nm, which could be interpreted as the high energy hybrid state resulting from strong coupling between the low energy LSPR with the SubPc Q

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band. In this strong coupling scenario, the Q band would therefore both provide the permittivity necessary to excite a dipolar resonance in the Ag nanoparticles, and at the same time strongly couple to this resonance to form hybrid states. However, further investigations are necessary to confirm the strong coupling scenario in this particular system.

4. Conclusions

In summary, we observed two bands from the absorption differences between thin-films consisting of a Ag NP layer covered by SubPc and pure SubPc thin-films. By means of numerical simulations, plasmonic absorption enhancement in the SubPc layer as well as absorption in the Ag NPs was found to contribute to both bands. By considering the simulated electric field surrounding the Ag NPs as well as calculations applying the quasi-static approximation, we were able to attribute the absorption in the Ag NPs at both absorption bands to dipole LSPRs. It should be further noted that the presence of multiple dipole resonances was not observed in the Ag NP layer without the SubPc medium surrounding it. Given this, we conclude that embedding spherical metal NPs in a highly dispersive medium can give rise to multiple absorption bands originating from dipole resonances, which is otherwise only exhibited by non-spherical NPs [30], and could be employed to broaden the spectral range of metal NP-based sensors. Furthermore, as the increase in absorption in SubPc benefits from the multiple LSPRs of the Ag NPs [Fig. 3(a)], this feature is expected to find application in organic solar cells, where enhanced absorption in the red tail of the absorption spectrum of the organic semiconductor should give rise to a corresponding increase in photocurrent [31,32].

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