Emission polarization dependence of strongly coupled modes between surface plasmon polaritons and dye molecules

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Nowadays surface plasmon polaritons (SPPs) coupled to optically active molecules are essential part of plasmonics. Many interesting effects involving coupled states are observed. Strong coupling with Rabi splitting up to several hundreds of meV is demonstrated [1,2]. Coupling from SPPs to far-field radiation via molecules is employed as SPP propagation imaging tool. Surface plasmon coupled emission provides high fluorescence detection sensitivity and spatial separation of surface and bulk generated fluorescence.

We have studied properties of the strongly coupled molecule-SPP (M-SPP) -polaritons via their emission polarization, and especially the effect of the Stokes shift of the molecule. The SPPs were excited using attenuated total refraction method [3]. Dye molecules possessing different Stokes shift (Nile Red, Rhodamine 6G and TDBC J-aggregates) were spin-coated within a polymer matrix on top of a 50 nm silver film. The dispersion relations were obtained and fitted by coupled oscillators model. We collected polariton emission/scattering with two different polarizations as a function of excitation angle. Samples without molecule layer showed only TM polarized emission, as assumed for pure SPP. However, when dye molecules were present, TE polarized signal also appeared. Thus, we can assign the TE emission to the properties of M-SPP-polariton. It was observed that close to the gap opening in the dispersion relation the TM/TE radiation ratio decreases, which can be explained by polariton mode turning more to a molecule like. Also an interesting observation is that the amount of TE radiation increases when Stokes shift of the dye decreases.

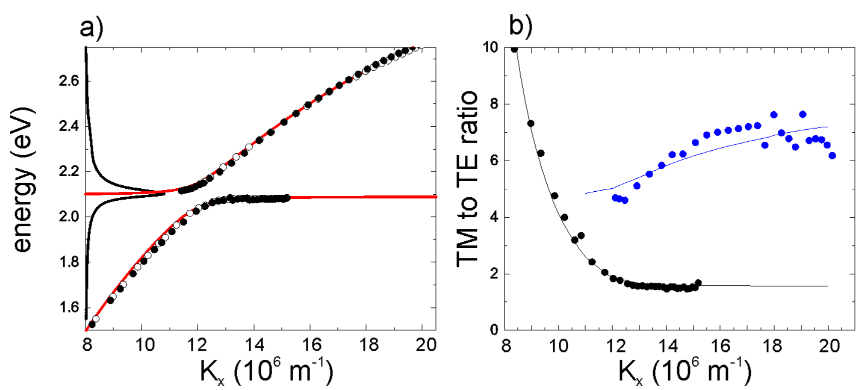

Figure 1: (a) Experimental dispersion curve for TDBC/silver sample (black and empty dots) together with coupled oscillator model fit (solid red line). (b) Ratios of TM to TE emission intensity for lower branch (black dots) and upper branch (blue dots). Theoretical fit (solid black and blue curves) is based on relative weights of SPP and molecule excitations.

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