Theory of absorption-induced transparency

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Abstract

Absorption induced transparency (AIT), roughly speaking, is a peak seen in the transmission spectrum of a holey metal film when a molecular dye is deposited on top of it [1]. The AIT peak appears unexpectedly close to one of the absorption energies of the molecules, hence its name. Tentative explanations of AIT pointed to strong-coupling interactions between Surface-Plasmon-Polaritons (SPPs) and molecules, when they are close by the metal surface (not inside the holes) [1].

Here we present our findings and extend our recent theoretical work on AIT [2]. We show the actual physical mechanism behind AIT. This takes place through a strong modification of the propagation constant of holes, $k_z$, so the holes must be at least partially filled. The spectral position of an AIT peak, its intensity and full width is mainly controlled by the spectral features of $k_z$. Therefore AIT has a localized character, which also explains that it occurs in single holes. In addition, we demonstrate that hole arrays in the AIT regime behave like a metamaterial characterized by a dielectric constant composed by a Drude plasma term (geometric origin) plus a Lorentz term due to the molecules. We also show that AIT peaks are nonplasmonic in character, so they are expected to occur at frequency regimes different than the optical, which opens the door for detection spectroscopy of chemical compounds with sharp absorption lines in the THz or microwave regimes.

To illustrate how AIT depends on the amount of molecular dye in the holes, we show in Fig.1(b) three transmission curves as a function of the wavelength, calculated with the Finite-Difference Time-Domain method (see caption for details). We choose the parameters in our model so the results best match the ones in Ref. [1], the dielectric constant of the molecules characterized by a Lorentz function. When the molecules are present and filling the holes (solid line) transmission clearly displays an AIT peak around 710nm, which is absent both without the molecules (dotted line) and even when they are not allowed to fill the holes, but deposited within a thin layer of polymer (dashed line). The rest of transmission features are Extraordinary Optical Transmission (EOT) peaks, boosted by the excitation of SPPs.

References

[1] J.A. Hutchison, D.M. O'Carroll, T. Schwartz, C. Genet, and T.W. Ebbesen, “Absorption-induced transparency”, Angew. Chem. Int. Ed. 50, 2085 (2011).
[2] S.G. Rodrigo, F.J. Garcia-Vidal, and L. Martín-Moreno, “Theory of absorption-induced transparency”, Phys. Rev. B 88, 155126 (2013).
Fig. 1: (a) An optically thick silver film is illuminated from the top, the absorption spectrum calculated then for two different “covering” media: air (dotted line) and molecules embedded within a polymer (solid line). (b) Transmission through a hole array covered and filled with molecules (solid line), just covered - no molecules inside- (dashed line) and without molecules (dotted line). Two EOT peaks are observed at wavelengths shorter than 650nm. The “filled” configuration shows an “extra” feature around 710nm, an AIT peak, apparently appearing at the absorption energy of the molecules. Hole array parameters: circular holes (radius = 70 nm), period $P = 250$ nm, and 200nm metal film thickness. The polymer layer is 30 nm in width.