Berreman mode and epsilon near zero mode

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Abstract: In this paper, we discuss the existence of an electromagnetic mode propagating in a thin dielectric film deposited on a metallic film at the particular frequency such that the dielectric permittivity vanishes. We discuss the remarkable properties of this mode in terms of extreme subwavelength mode confinment and its potential applications. We also discuss the link between this mode, the IR absorption peak on a thin dielectric film known as Berreman effect and the surface phonon polariton mode at the air/dielectric interface. Finally, we establish a connection with the polarization shift occuring in quantum wells.

OCIS codes: (240.6690) Surface waves; (310.6860) Thin films, optical properties; (260.3060) Infrared; (240.0310) Thin films.

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1. Introduction

It is well known that thin dielectric films deposited on top of a plane metallic surface modify the surface plasmon dispersion relation [1]. This is the basis of a very sensitive technique to detect the presence of biological material adsorbed on a surface [2]. This is however a slight perturbation of the surface plasmon mode. When studying the effect of a thin dielectric film in the infrared, the physics changes as the dielectric constant may become negative for frequencies between the longitudinal and the transverse optical phonon frequencies [3]. Hence, the dielectric-air interface can support a phonon-polariton surface mode and a stronger perturbation of the surface plasmon mode is expected. Free-standing dielectric films can support virtual modes (i.e. modes with a complex frequency and a real wavevector) [4] with particular properties at the Brewster angle or for phonon frequencies. Optical properties of dielectric thin films on a metal were studied in the past and absorption peaks, known as Berreman absorption peaks, have been reported [5] when studying the spectral reflectivity at a fixed angle. They have been attributed to the excitation of longitudinal modes in the early litterature [5–7]. This interpretation was later shown to be incorrect [8, 9]. Here, we revisit the analysis of the modes of a thin dielectric film deposited on a metal in the frequency range where the dielectric constant approaches zero. There has been a revival in the interest to materials with permittivity close to zero in recent years [10, 11]. In this paper, we are particularly interested in the possibility of ultra strong confinment of the field in a film with a thickness on the order of a few nanometers. This may have applications to design absorbers. It can also be extremely useful to enhance the coupling between electromagnetic fields and electrons if both are confined in a quantum well with a thickness of a few nanometers. In what follows, we search the modes of the structure. A specificity of our approach is that we account for retardation effects by searching modes with complex frequencies and real wavevectors parallel to the interface. Our analysis allows clarifying the origin of the Berreman absorption peak in terms of the excitation of a leaky mode that we call Berreman mode. It also provides a link between surface waves and the Berreman mode. We use the term surface wave to refer to waves corresponding to a pole $(\omega, K_\parallel)$ of the Fresnel factors lying beyond the light line. These were introduced by Sommerfeld in the context of radio waves and by Fano in the context of optics [12]. They are named surface plasmons polaritons for metals in the visible and surface phonon polaritons for dielectrics in the IR. We discuss in detail the structure of the mode for very thin dielectric slabs and introduce the concept of ENZ mode for wavevectors out of the light cone. Finally, we establish a connection with the so-called polarization shift observed in dopped quantum wells.

2. Complex frequency modal analysis

The geometry of the system is depicted in Fig. 1. A dielectric film with permittivity ε_2 and thickness d is deposited on a metal film with permittivity ε_3. The upper medium is characterized by a permittivity ε_1.

We search a TM mode in the form $H_y \exp(iK_\parallel x + ik_{z,1}z)$ in medium 1 and $H_y \exp(iK_\parallel x - ik_{z,3}z)$ in medium 3 where $k_{z,n} = (\varepsilon_n \omega^2/c^2 - K_\parallel^2)^{1/2}$ with the determination choice $\mathrm{Re}(k_{z,n}) + \mathrm{Im}(k_{z,n}) > 0$, and a time dependance in $\exp(-i\omega t)$. The dispersion relation of the TM mode is given by

$$\left(1 + \frac{\varepsilon_1 k_{z,1}}{\varepsilon_3 k_{z,3}}\right) = i \tan(k_{z,2} d) \left(\frac{\varepsilon_2 k_{z,3}}{\varepsilon_3 k_{z,2}} + \frac{\varepsilon_1 k_{z,2}}{\varepsilon_2 k_{z,1}}\right) \tag{1}$$

For lossy materials, this equation has no solutions for real values of the frequency and the wavevector. It is possible to find solutions by complexifying either the circular frequency ω or the wavevector $K_\parallel$. Finding a prescription for the choice is important as the resulting dispersion relations differ. This question has been discussed at length in Ref. [13]. The choice appears

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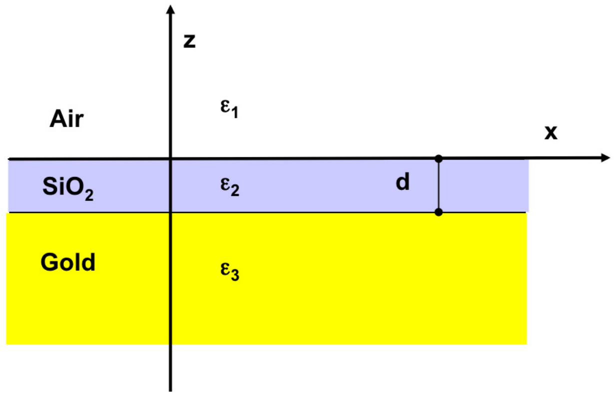

Fig. 1. Sketch of the geometry. A SiO_2 film is deposited onto a gold substrate. The thickness of this film is denoted by d. In the next figures, the upper medium is air ($\varepsilon_1 = 1$).

naturally when starting from a Fourier expansion of the Green tensor over real $K_\parallel$ values of the parallel wavevector and real values of the frequency, the z-component of the wavector being fixed by $k_{z,n} = (\varepsilon_n \omega^2/c^2 - K_\parallel^2)^{1/2}$. The Green tensor has poles which are given by Eq. (1). These poles correspond to modes of the system which can be either leaky modes, surface modes or guided modes. Using the residue theorem, it is possible to obtain the pole contribution by integrating analytically over either the frequency or the wavevector $K_\parallel$. Each representation is well suited to certain problems and corresponds to a given dispersion relation. For instance, integrating analytically over frequencies yields a field representation with real $K_\parallel$ and complex frequencies. This field representation allows discussing the density of states, the response of a system to a pulse illumination [13] and the steady state measurements of spectral responses at fixed angle as discussed in Ref. [14].

In order to solve the equation for complex frequencies, an analytical model of the dielectric constant is needed. Here, we use the following model for the silica permittivity in the range $950 - 1250\,\mathrm{cm}^{-1}$ ($1.79 - 2.36\,10^{14}\,\mathrm{rad.s}^{-1}$):

$$\varepsilon(\omega) = \varepsilon_\infty \frac{\omega^2 - \omega_L^2 + i\omega\Gamma}{\omega^2 - \omega_T^2 + i\omega\Gamma}, \tag{2}$$

where the constants have been obtained from a fit of the values reported in Ref. [15], $\varepsilon_\infty = 2.095$, $\omega_L = 1220.8\ \mathrm{cm}^{-1}$ ($2.30\,10^{14}\,\mathrm{rad.s}^{-1}$), $\omega_T = 1048,7\,\mathrm{cm}^{-1}$ ($1.98\,10^{14}\,\mathrm{rad.s}^{-1}$), $\Gamma = 71,4\,\mathrm{cm}^{-1}$ ($1.35\,10^{13}\,\mathrm{rad.s}^{-1}$). For a thickness of the dielectric film such that the two interfaces do not interact ($\mathrm{Im}(k_z)d >> 1$), we expect to find the dispersion relation of the surface phonon polariton at the air/dielectric interface and the dispersion relation of the surface plasmon at the metal/dielectric interface. This limit can be recovered in the limit $K_\parallel \to \infty$ so that $\tan(k_z d) \to i$, $k_z \to iK_\parallel$. The dispersion relation becomes:

$$(\varepsilon_1 + \varepsilon_2)\left(\frac{1}{\varepsilon_3} + \frac{1}{\varepsilon_2}\right) = 0, \tag{3}$$

so that we recover either the dispersion relation of the surface wave at the upper interface $\varepsilon_1 + \varepsilon_2 = 0$ or the dispersion relation at the lower interface $\varepsilon_2 + \varepsilon_3 = 0$. For a dielectric film on a metal, these solutions are respectively the surface phonon-polariton mode at the air-dielectric interface and the surface plasmon at the metal-dielectric interface. It follows that the dispersion relations are given by an asymptote for two fixed different frequencies. We now turn to

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the discussion of the dispersion relation for smaller values of the dielectric film thickness assuming that the upper medium is vacuum. We report a numerical study of the solutions of Eq. (1). Figure 2 shows the dispersion relation for three different values of the thickness of the dielectric film in an IR band close to the optical phonon frequency at $\omega_L = 1220.8\ \mathrm{cm}^{-1}$ ($2.30\,10^{14}\ \mathrm{rad.s}^{-1}$). We first pay attention to wavevectors in the light cone ($K_\parallel < \omega/c$). For a thin film, it is seen that a mode appears at a fixed frequency ω_L corresponding to the longitudinal optical frequency. Since the mode is in the light cone, it can leak in vacuum. It can be cheked that the Berreman absorption peak position in the $(\omega, K_\parallel)$ plane coincides with this dispersion relation for a thin film. Hence, we interpret the Berreman absorption peak as due to the excitation of this leaky mode. Note that the phenomenon of resonant absorption by a surface plasmon in Kretschman configuration can also be interpreted as the excitation of a leaky mode.

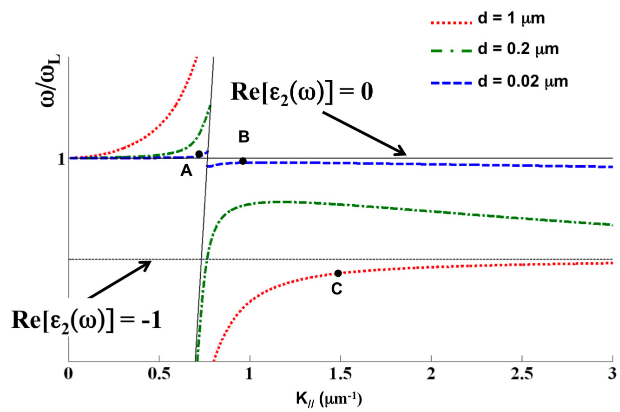

Fig. 2. Dispersion relation for three different film thicknesses. Point A corresponds to the Berreman leaky mode, Point B corresponds to the ENZ mode, Point C corresponds to the surface phonon polariton mode. The oblique thin line is the light line $\omega = cK_\parallel$. The two horizontal lines correspond to $\mathrm{Re}[\varepsilon_2(\omega)] = 0$ (close to ENZ) and $\mathrm{Re}[\varepsilon_2(\omega)] = -1$, which is the asymptot of the plane-interface surface-phonon-polariton dispersion relation.

Let us now consider the region out of the light cone ($K_\parallel > \omega/c$). As anticipated, for a large thickness (point C), the dispersion shows an asymptote close to $\varepsilon_2 + 1 = 0$ which is a clear signature of the surface phonon polariton at the air-dielectric interface. When reducing the thickness, the interaction between the two interfaces results in a shift of the mode frequency towards larger values. Finally, for a dielectric film much thinner than the wavelength, it is seen that the dispersion relation coincides with the horizontal line corresponding to a frequency such that $\Re(\varepsilon(\omega)) \to 0$ as indicated in the figure. It is easy to understand this behaviour by realizing that the surface wave at the air/dielectric interface can exist provided that the real part of the dielectric constant remains negative. Hence, as the thickness decreases, the mode frequency increases but has to remain smaller than ω_L. The dispersion relation tends therefore towards the asymptote $\omega = \omega_L$.

We now study the structure of the modes obtained close to the longitudinal optical frequency. Figure 3(a) shows the form of the magnetic field and the z-component of the electric fied for the Berreman leaky mode. Note that the magnetic field increases in the vacuum which is a characteristic feature of a leaky mode. The right panel shows the highly localized electric field in the dielectric film. This confinement can be easily understood using a simple argument. The z-component of $\mathbf{D} = \varepsilon\mathbf{E}$ is continuous so that $E_{z,2} = (\varepsilon_1/\varepsilon_2)E_{z,1}$. It follows that the field $E_{z,2}$ is enhanced if ε_2 approaches zero. This enhancement of the field is the physical origin of the Berreman absorption peak. The second line of Fig. 3 shows the fields for point B, i.e. for a frequency close to ω_L and a wavevector larger than ω/c. The magnetic field decays away from the dielectric film while the electric field is both enhanced and confined in the film due to the vanishing dielectric constant. We thus call this mode ENZ mode. This mode is characterized by $\omega \approx \omega_L$ and $K_\parallel > \omega/c$, a flat dispersion relation so that the group velocity is much smaller than c and the density of states is very large and finally a confinment of the energy in a very thin slab. To quantify the field enhancement, we introduce an ENZ factor $K_{\mathrm{ENZ}} = |E_{z,2}/E_{z,1}|^2 = |\varepsilon_1/\varepsilon_2|^2$. We plot this factor as a function of frequency in Fig. 4. The enhancement is modest for amorphous silica as seen in Fig. 4. However, when using crystalline materials, it can become greater than 100.

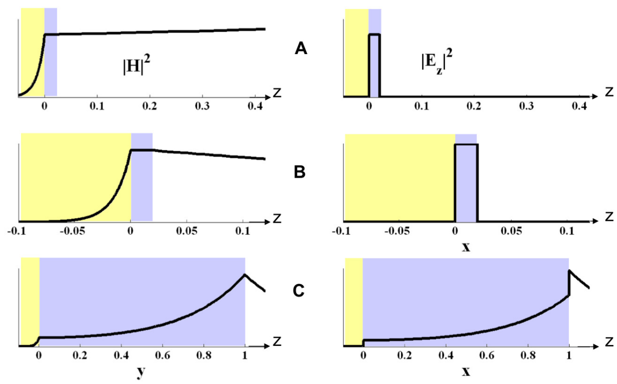

Fig. 3. Structure of the fields $|H|^2$ and $|E_z|^2$ (arbitrary units) for the Berreman mode (point A in Fig. 3), the ENZ mode (B), the surface phonon polariton mode (C). The layers are characterized by the same colors as Fig. 1.

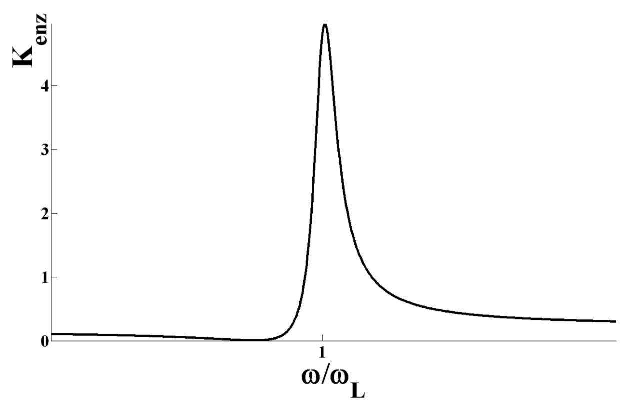

Fig. 4. ENZ enhancement factor

It is interesting to note that a similar phenomenon can be observed in quantum wells (QW). In this case, ε_1 and ε_3 are the barrier and ε_2 the QW dielectric constants. Here, the zero of the dielectric permittivity can be provided by the intersub-band transitions in the quantum well at a frequency which can be adjusted by changing the QW thickness. The modes of this system are also described by Eq. (1) so that a ENZ mode can exist in the QW. Due to the ENZ enhancement, the IR absorption peak is shifted from the intersub-band transition towards the frequency where the real part of the dielectric constant vanishes [16]. This effect is known as depolarization shift. This effect may be of particular interest for optoelectronics applications

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as this system allows confining simultaneously the electrons and the electromagnetic field in a very thin slab. Furthermore, the ENZ effect enhances the normal component of the electric field which interacts with the intersub-band transitions. It is thus promising to combine the ENZ enhancement due to phonons with a quantum well. This paves the way towards active systems as the QW electronic density can be tuned by applying a gate voltage. An example of electrically modulated reflectivity based on this idea will be reported elsewhere.

3. Analytical approach

Finally, we solve Eq. (1) analytically for frequencies approaching ω_L. It is interesting to derive an explicit form of the dispersion relation which is valid for both the Berreman leaky mode and the ENZ mode in order to exhibit the common origin of these two modes. Upon examination of Eq. (1), it is seen that for frequencies close to ω_L, the leading term on the right hand side varies as $1/\varepsilon_2$. We introduce the quantity $d' = \tan(k_{z,2}d)/k_{z,2}$ that tends to d when $d \to 0$. After rearranging terms, Eq. (1) can be cast in the form :

$$\omega = \omega_{\mathrm{ENZ}} + \frac{K_\parallel^2(\omega_L^2 - \omega_T^2)}{[A(\omega, K_\parallel) - K_\parallel^2][\omega + \omega_L^2/\omega_{\mathrm{ENZ}}]}, \tag{4}$$

where $\omega_{\mathrm{ENZ}} = \omega_L\sqrt{1 - \frac{\Gamma^2}{4\omega_L^2}} - i\frac{\Gamma}{2}$ is the complex solution of $\omega^2 - \omega_L^2 + i\omega\Gamma = 0$ (e.g. $\varepsilon_2(\omega) = 0$), and

$$A(\omega, K_\parallel) = \frac{i\varepsilon_\infty}{d'}\left[\frac{k_{z,1}}{\varepsilon_1} + \frac{k_{z,3}}{\varepsilon_3} - id'\left(\frac{\omega^2}{c^2} + \varepsilon_2\frac{k_{z,1}}{\varepsilon_1}\frac{k_{z,3}}{\varepsilon_3}\right)\right].$$

It is straightforward to obtain an iterative solution by replacing ω by ω_{ENZ} in the right hand side of Eq. (4). Figure 5 displays a comparison of the first order iterative solution with the numerical solution showing a remarkable agreement for small values of the thickness.

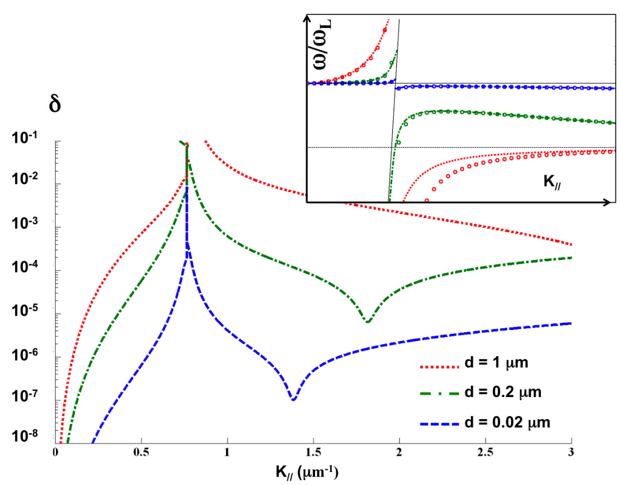

Fig. 5. Relative error (δ) between the approximate and exact solutions of the dispersion relation in logarithmic scale as a function of $K_\parallel$ for different thicknesses of the film. The inset presents the dispersion relation of Fig. 2, on which first order iterative solutions have been added (circles).

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4. Conclusion

In summary, we have shown that a thin dielectric film supports a leaky mode in the light cone and a surface mode beyond the light cone characterized by a complex frequency and a real wavevector. We attribute the Berreman absorption peak to the excitation of this leaky mode which we therefore call Berreman mode. Given the remarkable enhancement of the field in the dielectric film due to the vanishing amplitude of the dielectric constant at the longitudinal frequency, we call the surface mode ENZ mode. We anticipate that the ENZ mode can be resonantly excited through a grating and produce total absorption. When using semiconductors such as GaAs/AlGaAs, this type of system should lead to an extremly strong interaction between electrons and electromagnetic field. This paves the way towards new applications for optoelectronic devices in the IR at ambiant temperatures. The study of these effects will be reported in future publications.

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