Some considerations on the transmissivity of thin metal films

Ian R. Hooper and J. Roy Sambles*

School of Physics, University of Exeter, Exeter, Devon, EX4 4QL, UK
Corresponding author: j.r.sambles@exeter.ac.uk

Abstract: As interest in plasmonics grows the optical properties of thin metal films becomes increasingly significant. Here we explore the transmissivity of thin metal films at normal incidence, from the ultraviolet to microwaves, and show how, contrary to simplistic treatments, the microwave transmissivity may be much less than the optical transmissivity for films which are well below the skin depth in thickness. This arises because the film is acting as a zero order Fabry-Perot with very high reflectivity at each interface. The skin depth then becomes irrelevant for thin metal films at microwave frequencies. We also note in passing that the expected exponential dependence on thickness at higher thicknesses has an asymptotic limit at zero thickness which may be as high as four times the input intensity.

OCIS codes: (310.6860) Thin Films; (160.3900) Metals

References and links

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The optical properties of thin metal films has long been a subject of scientific research. Over the past decade there has been a resurgence of interest in this area primarily as a result of Ebbesen and coworkers observation of strongly enhanced transmission through holey metal films [1]. Further interest in thin metal films has also been stimulated by Pendry's suggestion of using thin metal layers as perfect lenses [2] while at much the same time the same author [3] has re-stimulated general interest in the idea of negative index materials – metals, with negative permittivities structured to give also negative permeabilities then lead consequentially to the possibility of 'cloaking' [4]. It is in this context of renewed interest in the optical properties of structured metal films that we here revisit the rather old problem of the transmissivity of unstructured films.

There are various standard texts which give expressions for the transmissivity of a thin slab of any material. Stratton [5] gives the transmissivity T (Stratton p515 Eq. (24)) for normal incidence in air as:

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$$T = \frac{(1-R)^2 + 4R\sin^2\delta}{\left(e^{\beta d} - \mathrm{Re}^{-\beta d}\right) + 4R\sin^2(\delta + \alpha d)}$$

with $\alpha = (\omega / c) n_m = n_m k_0$, $\beta = (\omega / c) k_m = k_m k_0$ (the refractive index of the metal is $n_m + ik_m$, the thickness is d and k_0 is the free-space wavevector of the light). Also,

$$R = \frac{(1-n_m)^2 + k_m^2}{(1+n_m)^2 + k_m^2} \qquad \text{(Stratton p 512 Eq. (12))}$$

and

$$\tan\delta = \frac{2k_0\beta}{\alpha^2 + \beta^2 - k_0^{\,2}} = \frac{\sqrt{2}\left(-\varepsilon_r + \left(\varepsilon_r^2 + \varepsilon_i^2\right)^{1/2}\right)^{1/2}}{\left(\varepsilon_r^2 + \varepsilon_i^2\right)^{1/2} - 1} \qquad \text{(Stratton p513 Eq. (15))}$$

where $\varepsilon_r = n_m^2 - k_m^2$ and $\varepsilon_i = 2n_m k_m$, where the subscripts *r* and *i* correspond to the real and imaginary parts (as they do throughout this text).

Another version is found in Reitz, Milford and Christy [6]. Beginning from the transmission coefficient of amplitude (Ref. 6 p. 464 Eq. (18.86))

$$t = \frac{t_{12}t_{21}}{\left(e^{-i\beta'/2} + r_{12}r_{21}e^{i\beta'/2}\right)} = \frac{\left(1 - r_{12}^2\right)}{\left(e^{-i\beta'/2} - r_{12}^2 e^{i\beta'/2}\right)}$$

then *T* takes the form

$$T = \frac{(1-A)(1-A^*)}{\left(e^{-i\beta'/2} - Ae^{i\beta'/2}\right)\left(e^{i\beta'^*/2} - A^* e^{-i\beta'^*/2}\right)}$$

with $\beta' = 2dk_0(n_m + ik_m)$ and $A = \left[\dfrac{n_m - 1 + ik_m}{n_m + 1 + ik_m}\right]^2$

for which $A_r = \dfrac{\left(n_m^2 + k_m^2 - 1\right)^2 - 4k_m^2}{\left(n_m^2 + k_m^2 + 2n_m + 1\right)^2}$ and $A_i = \dfrac{4k_m\left(n_m^2 + k_m^2 - 1\right)}{\left(n_m^2 + k_m^2 + 2n_m + 1\right)^2}$.

These two seemingly rather different expressions for T are readily found to be equivalent using the substitutions: $A_r = R\cos(\delta/2)$ and $A_i = R\sin(\delta/2)$. Such expressions, and their more complex off-normal forms, can of course be readily written into a computer code and it is a simple matter then to compute the transmissivity of any metal film at any angle of incidence.

To illustrate the interesting results which may be forthcoming from using such expressions we plot in Fig. 1 the transmittance as a function of sample thickness for silver films modeled with a Drude approximation having ω_p = 1.32 x $10^{16}\mathrm{s}^{-1}$ (k_p=ω_p/*c*) as the plasma frequency and τ = 1.45 x 10^{-14}s the relaxation time, for which:

$$\varepsilon_r = 1 - \frac{(\omega_p\tau)^2}{1 + (\omega\tau)^2} \text{ and } \varepsilon_i = \frac{\omega_p^{\,2}\tau}{\omega\left(1 + (\omega\tau)^2\right)}.$$

Figure 1(a) shows this modeling in linear form while Fig. 1(b) illustrates it in logarithmic fashion.

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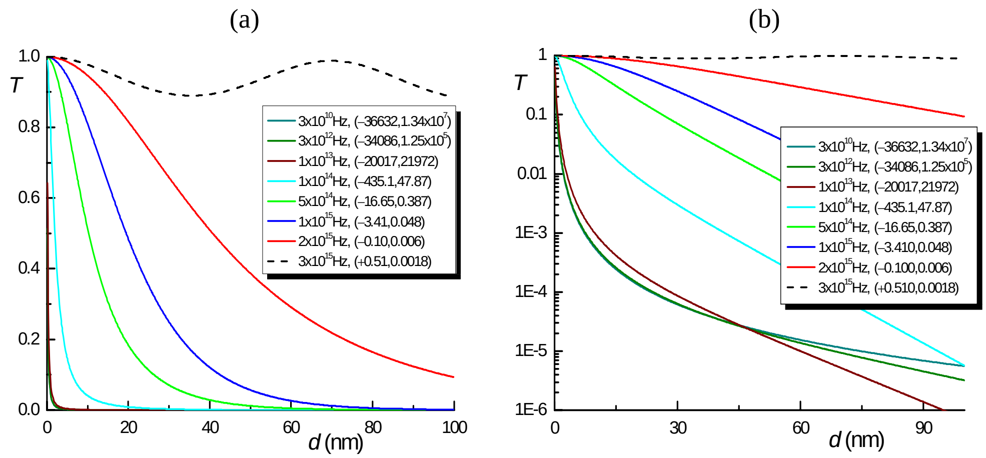

Fig. 1. Plots of the transmissivity T as a function of silver film thickness *d* for a wide range of frequencies (permittivities in brackets). (a). linear; (b). logarithmic (Note that the line for $f = 3 \times 10^{15}$ Hz corresponds to light above the plasma frequency and as such the silver is no longer acting as a metal – it is added for comparison only).

These figures illustrate some obvious key points. Firstly, there is a clear oscillation evident in the high frequency data set ($f = 3\text{x}10^{15}$Hz). At this frequency the film is acting as a dielectric with the oscillations arising from interference between reflections from the front and back interfaces of the slab. This data set is only plotted here for comparative purposes; the important points to note are those arising from the regime in which the material of the slab is acting as a metal. Secondly, for thicknesses above approximately 60 nm, regardless of frequency, a silver film allows less than a few percent of light to be transmitted. Thirdly, for all thicknesses above about 40 nm the thickness dependence is exponential, as expected, seemingly dominated in form by the skin depth (we shall return to this later). Finally, as is most apparent in Fig. 1(b), at microwave frequencies the initial thickness dependence is far from exponential and indeed much faster than for shorter wavelengths. Further, by 80 nm in thickness the transmissivity for microwaves is greater than for shorter wavelengths, although in reality by that thickness both are almost negligible ($<10^{-5}$).

In view of the unexpected thickness dependence at microwave frequencies and in order to explore further the thickness dependence of the transmissivity we look to express this transmissivity as a function of film thickness.

First it is helpful to re-express T as:

$$T = \frac{(1-A_r)^2 + A_i^2}{\left(A_r^2 + A_i^2\right)e^{-2dk_m k_0} + e^{2dk_m k_0} - 2A_r\cos(2dn_m k_0) + 2A_i\sin(2dn_m k_0)}$$

For high thicknesses it is apparent that the dominant term will be simply:

$$T \approx \left[(1-A_r)^2 + A_i^2\right]e^{-2dk_m k_0} \tag{1}$$

Then, on a logarithmic plot, we find ln T has an intercept given by $\ln\left[(1-A_r)^2 + A_i^2\right]$ and a slope of $-2k_m k_0$. Note the presence of the intercept, the thickness dependence is not just exponential with an origin at $T = 1$. This illustrates the danger of simply treating the thickness dependence of the transmissivity of a metal film as an exponential, since extrapolating to zero thickness could readily yield an apparently nonsensical result. Examination of Fig. 1 for

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frequencies above 5 x 10^{14} Hz illustrates this point fully. The model data will fit a straight line with an intercept beyond 1! The transmission at large d can easily be described by $t_{12}t_{23}\exp(-k_m k_0 d)t_{12}^* t_{23}^* \exp(-k_m^* k_0 d)$ because we can neglect multiple reflections. In other words the field on the transmission side of the first interface (t_{12}) is attenuated through the film ($\exp(-k_m k_0 d)$) before being transmitted through the second interface (t_{23}). The assumption that the intercept of the exponential should be less than 1 comes from the assumption that the field on the transmission side of the first interface will be less than 1. This is not the case. Consider frustrated total reflection in the case of two prisms close together with light incident beyond the critical angle. When they are far apart the evanescent field amplitude just beyond the interface is twice the input amplitude. When the second prism is introduced within the tail of the evanescent field this value of 2 at the first interface will only change when the influence of multiple reflections within the air gap are taken into account. Consequentially the exponential part of the transmission as a function of the gap may have an intercept greater than 1. The maximum this may be occurs when $n_m = 0$, $k_m = 1$ so that $t_{12} = 1-i$ and $t_{23} = 1+i$. Hence $t_{12}t_{23} = 2$ and the form of the exponential decay of the transmitted intensity becomes $(2\exp(-k_0 d))^2$ with an intercept at $d = 0$ of 4.

In addition, the varying gradient at large thickness evident in Fig. 1(b) can also be explained via inspection of Eq. (1), from which the slope at large thickness is expected to be equal to $-2k_m k_0$. How does this fit with the data in Fig. 1(b), where for long wavelengths the transmission through a thicker film can be larger than for shorter wavelengths? In Fig. 2 the value of $-2k_m k_0$ is plotted as a function of frequency.

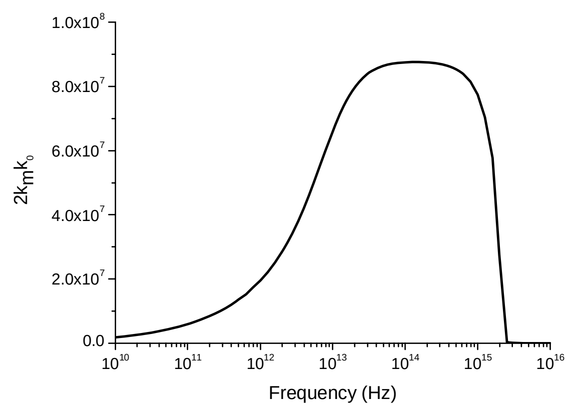

Fig. 2. $2k_m k_0$ as a function of frequency with k_m being calculated using the values of ω_p and τ as defined in the text.

It is evident from Fig. 2 that the gradients of the transmission plots in Fig. 1(b) at large thickness values are expected to vary depending upon the frequency of the light. The greatest gradient occurs for a frequency range of 10^{14}-10^{15}Hz, with lower gradients for both higher and lower frequencies. The much shallower gradient for low frequencies as compared to those in the 10^{14}-10^{15}Hz range results in a higher transmission at large film thickness. This is due to the particular form of the dispersion of k_m with changing frequency.

However we are here rather more concerned with lower thicknesses. The above general expression may be rewritten as:

$$\frac{(1-A_r)^2 + A_i^2}{T} = \left(A_r^2 + A_i^2\right)e^{-2dk_m k_0} + e^{2dk_m k_0} - 2A_r\cos(2dn_m k_0) + 2A_i\sin(2dn_m k_0)$$

which may be expanded in orders of d. To lowest order ($dk_m k_0 \ll 1$):

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$$\frac{(1-A_r)^2 + A_i^2}{T} \cong \left(A_r^2 + A_i^2\right) + 1 - 2A_r$$

which leads directly to $T = 1$, not a particularly illuminating result. To the next order:

$$\frac{(1-A_r)^2 + A_i^2}{T} \cong \left(A_r^2 + A_i^2\right)(1 - 2dk_m k_0) + (1 + 2dk_m k_0) - 2A_r + 2A_i(2dn_m k_0)$$

which is best re-expressed as:

$$\frac{1-T}{T} \cong 2dk_0\left[\frac{k_m\left(1 - A_r^2 - A_i^2\right) + 2A_i n_m}{(1-A_r)^2 + A_i^2}\right].$$

Substituting in for A_r and A_i from above one finds that

$$\left[\frac{k_m\left(1 - A_r^2 - A_i^2\right) + 2A_i n_m}{(1-A_r)^2 + A_i^2}\right] = \frac{\varepsilon_i}{2}.$$

Thus the final expression for the transmissivity of a very thin metal film is, to first order, $(1-T)/T \cong \varepsilon_i dk_0$. Note that to this approximation the transmissivity is entirely dominated by ε_i and is linear in d.

How good an approximation is this? Is it of any use over a broad spectral range? To illustrate this the calculated transmissivity of thin silver and aluminium films at 632.8 nm are examined.

The graph in Fig. 3 shows T for silver and aluminium as a function of d for a full numerical calculation based on the 3 layer Fresnel model and for successive higher order approximations (see below) using experimental values for ε_m.

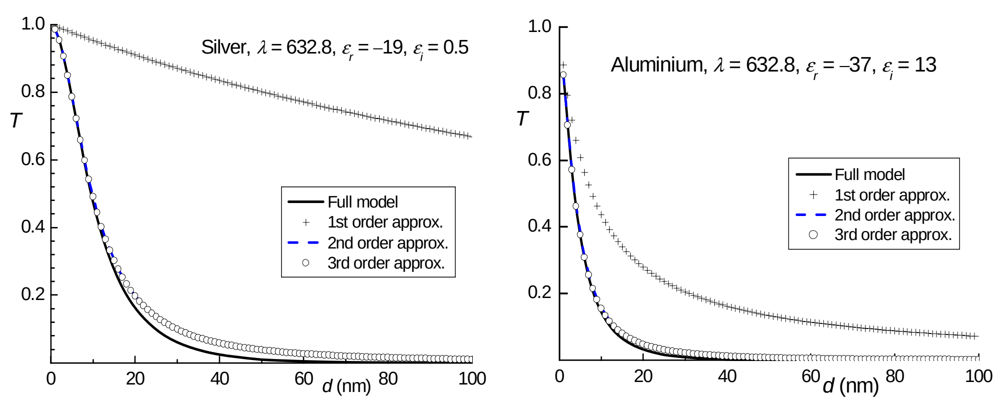

Fig. 3. *T*ransmission as a function of film thickness d for silver and aluminium at 632.8 nm. For silver $\varepsilon_m = -19+0.5i$ [7] and for aluminium $-37+13i$ [8].

It is immediately apparent that for silver the first order approximation is very poor but that in the case of aluminium, with a much larger ε_i, it is far better. This is, of course, as expected for the first order approximation since it only contains ε_i and as such any material which has a

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larger ratio of ε_i to ε_r will obey the approximation more closely. Also note that aluminium has a much lower transmissivity than silver at this wavelength of 632.8 nm.

It is worthwhile noting what happens if a Drude model is used to extend this type of calculation far beyond the visible. This is shown in Fig. 4 for silver films of 20 and 40 nm thickness with Drude parameters $\omega_p = 1.32 \times 10^{16}$ and $\tau = 1.45 \times 10^{-14}$.

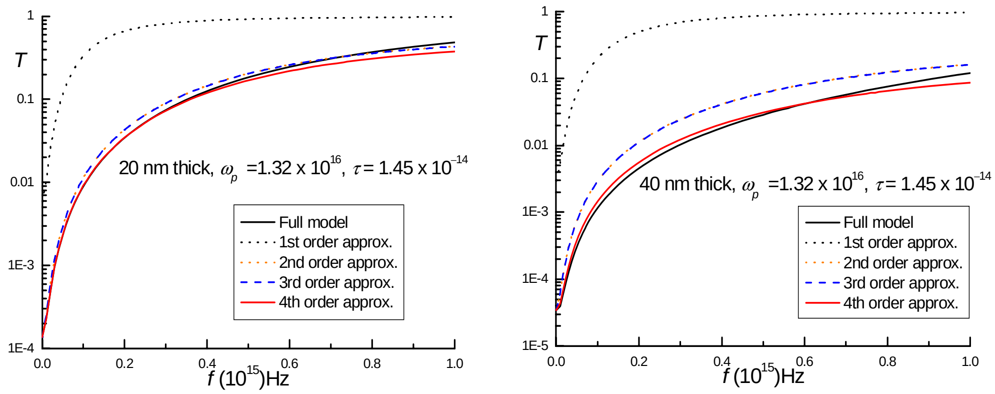

Fig. 4. First order calculation for transmissivity T, using the Drude model for silver, with $\omega_p = 1.32 \times 10^{16}\ s^{-1}$ and $\tau = 1.45 \times 10^{-14}$ s for d = 20 nm and d = 40nm. The solid line is exact.

Note that the transmissivity is very small for all longer wavelengths, even though the metal films are only 20 nm and 40 nm thick. These thicknesses are very much less than the skin depths at these longer wavelengths. Indeed, the transmissivity at these low thicknesses falls far more rapidly with increasing thickness than a skin depth model would suggest. This is due to the enormous impedance mismatch between the air and the metal at these longer wavelengths where $\varepsilon_i >> 1$ and the very strong reflectivity at both interfaces results in the metal layer acting as a zero order Fabry-Perot resonator. Consequently the very weak transmission one sees at small thicknesses for long wavelengths has little directly to do with the skin depth effect but rather reflects the thickness dependence of the multiple interference of the radiation within the metal layer.

The general series expansion for the transmissivity is of the form:

$$\frac{1-T}{T} = \sum_{odd\ p} \frac{1}{p!}(2dk_0)^p \left[\frac{k_m^p\left(1-A_r^2-A_i^2\right)+2A_i n_m^p(-1)^{\left(\frac{p-1}{2}\right)}}{(1-A_r)^2+A_i^2} \right] + \sum_{even\ p} \frac{1}{p!}(2dk_0)^p \left[\frac{k_m^p\left(1+A_r^2+A_i^2\right)+2A_r n_m^p(-1)^{\left(\frac{p-2}{2}\right)}}{(1-A_r)^2+A_i^2} \right]$$

This gives, to fourth order:

$$\frac{1-T}{T} \cong \varepsilon_i(dk_0) + \frac{\varepsilon_r^2+\varepsilon_i^2-2\varepsilon_r+1}{4}(dk_0)^2 + \frac{\varepsilon_i}{3}(1-\varepsilon_r)(dk_0)^3 + \frac{2\varepsilon_r^2+2\varepsilon_i^2-\varepsilon_r\left(\varepsilon_r^2+\varepsilon_i^2\right)-\varepsilon_r}{12}(dk_0)^4$$

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Analytic forms for the Drude model

For a metal which follows the Drude approximation, and neglecting the background ionic contribution at low frequencies where this will be negligible, one may define:

$$\varepsilon_A^2 = \varepsilon_r^2 + \varepsilon_i^2 = \frac{(\omega_p \tau)^4}{\left(1+(\omega\tau)^2\right)^2} + \frac{\omega_p{}^4 \tau^2}{\omega^2 \left(1+(\omega\tau)^2\right)^2} = \frac{\omega_p{}^4 \tau^2}{\omega^2 \left(1+(\omega\tau)^2\right)}.$$

By substitution into the fourth order expansion, this gives:

$$\frac{1-T}{T} \cong \frac{\omega_p{}^2 \tau}{\omega\left(1+(\omega\tau)^2\right)}(dk_0) + \frac{1}{4}(dk_0)^2\left[\frac{\omega_p{}^4\tau^2}{\omega^2\left(1+(\omega\tau)^2\right)} + 2\frac{(\omega_p\tau)^2}{1+(\omega\tau)^2} + 1\right]$$

$$+\frac{1}{3}(dk_0)^3\left[1+\frac{(\omega_p\tau)^2}{1+(\omega\tau)^2}\right]\frac{\omega_p{}^2\tau}{\omega\left(1+(\omega\tau)^2\right)} + \frac{1}{12}(dk_0)^4\left[\left(2+\frac{(\omega_p\tau)^2}{1+(\omega\tau)^2}\right)\frac{\omega_p{}^4\tau^2}{\omega^2\left(1+(\omega\tau)^2\right)} + \frac{(\omega_p\tau)^2}{1+(\omega\tau)^2}\right]$$

which simplifies, for $\omega_p \tau >> 1$, to:

$$\frac{1-T}{T}\left(1+(\omega\tau)^2\right) \cong \omega_p\tau(dk_p) + \frac{1}{4}(dk_p)^2\left[(\omega_p\tau)^2 + 2(\omega\tau)^2\right]$$

$$+\frac{1}{3}(dk_p)^3(\omega_p\tau)\frac{(\omega\tau)^2}{\left(1+(\omega\tau)^2\right)} + \frac{1}{12}(dk_p)^4\left[(\omega_p\tau)^2\frac{(\omega\tau)^2}{\left(1+(\omega\tau)^2\right)} + \left(\frac{\omega}{\omega_p}\right)^2(\omega\tau)^2\right]$$

In the limit of relatively low frequencies, $\omega\tau << 1$, this expression further reduces to give:

$$\frac{1-T}{T} \cong \omega_p\tau(dk_p) + \frac{1}{4}(dk_p)^2(\omega_p\tau)^2.$$

The dominant term is in general the quadratic term in *d*, unless *d* or τ are very small. Notice that *T* is now independent of frequency, only depending on the material parameters. If now for this limit situation (microwave frequencies and below), 1/T is plotted against d^2 an extremely good straight line is obtained. This is illustrated in Fig. 5.

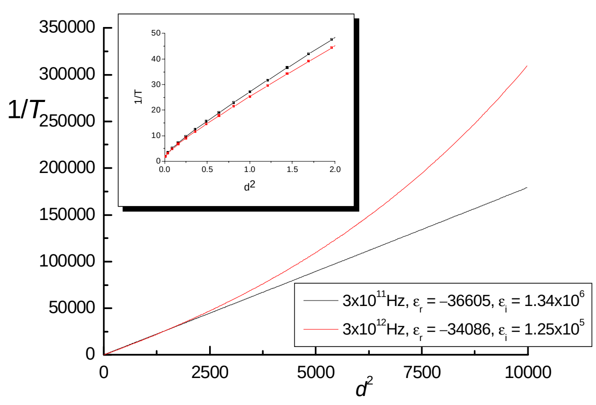

Fig. 5. Plot of 1/*T* against d^2 for silver at $f = 3\text{x}10^{11}$ and $3\text{x}10^{12}$Hz (inset: close up for low values of *d*). All plots for lower frequencies overlie that of the line for $f = 3\text{x}10^{11}$.

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It is very obvious from Fig. 5 that this second order approximation predicts very well the thickness dependence of the inverse transmissivity at low thicknesses at microwave frequencies and below (black line). Of course this is in reality of very little interest since the transmissivity is itself so low. However we can take this approximation to even lower thicknesses as shown in the inset of Fig. 5. Note that this simple quadratic relationship fails for the very lowest thicknesses (unrealistically less than 1 nm) as a contribution from the linear term begins to have an effect. The red line corresponds to THz frequencies where the approximation has begun to fail and other terms begin to take effect. At higher frequencies this becomes even more pronounced. (It should be noted however, that in the low thickness regime the approximation still agrees well).

So what is the fundamental physical cause of this d^2 dependence? Why, at microwave frequencies, is the simple exponential expected from skin depth considerations not manifest at these low thicknesses? The first thing to note is that the refractive index at these frequencies is very large, with equal magnitude real and imaginary components. The imaginary part governs the exponential thickness dependence at large thicknesses but for very thin films (much less than the wavelength and skin depth within the metal) this exponential term is insignificant. The primary effect in this regime is the interference of the strong reflections from the front and back faces of the thin metal plate. The impedance mismatch with air is so large that to first order the reflection coefficient at each interface is 1. Thus the film is a high Q Fabry-Perot resonator, of zero order. Generally one expects the cavity of a Fabry-Perot to have a thickness of order the external wavelength divided by twice the refractive index of the material. However this is for the first order mode, which at microwave frequencies (wavelength ~10 mm) with the real part of the metal refractive index of order 10^3 to 10^4, gives a required thickness of microns (typical of the skin depth thickness). Here one is dealing with much thinner layers, the zero thickness limit, or the zero order Fabry-Perot mode. Then, as the thickness is increased from zero ($T = 1$) the transmittance falls rapidly with d, as described by the equations given above, following the behaviour expected of a high Q Fabry-Perot cavity.

Returning to the starting equation one has:

$$t = \frac{t_{12}t_{21}}{\left(e^{-i\beta'/2} + r_{12}r_{21}e^{i\beta'/2}\right)} = \frac{\left(1 - r_{12}^2\right)}{\left(e^{-i\beta'/2} - r_{12}^2 e^{i\beta'/2}\right)}.$$

After substituting in for r_{12} and β this may be reexpressed as:

$$\frac{1}{t} = \left[\frac{1}{2} - \frac{1+\varepsilon_m}{4(n_m + ik_m)}\right]e^{i(n_m+ik_m)k_0 d} + \left[\frac{1}{2} + \frac{1+\varepsilon_m}{4(n_m + ik_m)}\right]e^{-i(n_m+ik_m)k_0 d},$$

From this the key d dependent term is:

$$-i\frac{\varepsilon_m}{2(n_m + ik_m)}\sin(n_m + ik_m)k_0 d \,.$$

This may be perhaps more helpfully written as

$$-i\left[\frac{\sin(\beta'/2)}{\beta'/2}\right]\varepsilon_m\left(\frac{k_0 d}{2}\right)$$

the typical sinc like behaviour. Now one can see that as β' tends to 0 so, for large ε_i, the simple $\varepsilon_i k_0 d/2$ dependence in $1/t$ will arise, leading in turn to $T \approx 1 - \varepsilon_i k_0 d$. Finally, returning to the general expression note that the fourth order term is likely to be the next order correction through the term $(1/12)(dk_p)^4(\omega_p\tau)^2(\omega\tau)^2$. This however contains an ω^2 term which means that it will be insignificant - as will all other higher order terms. This explains why the second order expression is such a good approximation in the low frequency limit.

Another route to the simpler expressions is to appreciate that in the long wavelength limit ε_i will dominate all other terms. This leads from the general quadratic to

$$\frac{1-T}{T} \cong \varepsilon_i(dk_0) + \frac{\varepsilon_i^2}{4}(dk_0)^2 - \frac{\varepsilon_i}{3}\varepsilon_r(dk_0)^3 - \frac{\varepsilon_r\varepsilon_i^2}{12}(dk_0)^4$$
$$= \varepsilon_i(dk_0)\left(1 - \frac{\varepsilon_r}{3}(dk_0)^2\right)\left(1 + \frac{\varepsilon_i}{4}(dk_0)\right)$$

Then using the substitutions $\varepsilon_r \approx -(\omega_p\tau)^2$ and $\varepsilon_i \approx \omega_p^2\tau/\omega$ in this expression leads to:

$$\frac{1-T}{T} \cong \frac{\omega_p{}^2\tau}{\omega}(dk_0)\left(1 + \frac{(\omega_p\tau)^2}{3}(dk_0)^2\right)\left(1 + \frac{\omega_p{}^2\tau}{4\omega}(dk_0)\right)$$
$$= \omega_p\tau(dk_p)\left(1 + \frac{(\omega\tau)^2}{3}(dk_p)^2\right)\left(1 + \frac{\omega_p\tau}{4}(dk_p)\right)$$

Which is of course identical to our previous approximation.

Conclusions

It is often naively assumed that the thickness dependence of the transmission of light through metal films is a simple exponential dictated by the skin depth. While for high thicknesses (for which the transmittance is actually often negligible) this is certainly true, it is generally not true for thicknesses relevant to many experimental thin film situations. Furthermore when it is true the extrapolated zero thickness value may well be greater than unity. Significantly, for the visible domain, there may be a substantially enhanced transmission, above that naively expected for 'good' metals such as silver, while in the microwave domain the transmittance for thin samples is governed not by the skin effect but by the metal film acting as a zero order Fabry-Perot resonator. The consequence is that, somewhat surprisingly, thin metal films, even at normal incidence, readily transmit a higher percentage of visible than of microwave radiation.

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