



# Nonlinear dispersion relation for surface plasmon at a metal–Kerr medium interface

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## ABSTRACT

The dispersion relation for the plasmon at the interface of a metal and a nonlinear Kerr medium is studied by applying a semi-analytic theory established in the literature. Explicit analytical results are obtained and are compared to those from a certain approximate treatment appeared in the literature. It is found that for large electric field strengths, both the dispersion relation and the surface plasmon frequency from the approximate treatment deviate significantly from those obtained in the exact approach, especially for the case with a negative Kerr susceptibility.

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## 1. Introduction

Surface plasmon (SP) excitation refers to the collective oscillation of the free electrons at a metal–dielectric interface with excitation characteristics depending on the optical properties of the metal and the dielectric, and on the geometry of the boundary interface between the two media. While it was first observed accidentally more than a century ago and its physics was clarified in the 1950s [1], the recent surge of the field of plasmonics [2] has revitalized the excitement of this subject leading to unprecedented activities in both fundamental and applied research in this emerging field of nano-optics.

In this communication, we study the excitation of SP at the interface of the simplest geometry – an infinite planar interface separating the metal and a dielectric, but allowing the dielectric to have nonlinear optical properties in general. In particular, we shall consider a Kerr-type dielectric with its dielectric function depending on the intensity of the electric field. While it is well-known that the dispersion for the SP (with wave number  $k$  and angular frequency  $\omega$ ) at a planar interface between a linear optical medium and a metal is given by the following expression [1]:

$$k = \frac{\omega}{c} \sqrt{\frac{\varepsilon_d \varepsilon_m}{\varepsilon_d + \varepsilon_m}}, \quad (1)$$

where  $c$  is the speed of light in vacuum and the dielectric functions of the dielectric ( $\varepsilon_d$ ) and metal ( $\varepsilon_m$ ) can depend only on the frequency; the case for a nonlinear dielectric–metal interface has also

been studied intensively in the literature for over three decades, especially for the case of a Kerr dielectric where it was found that the SP in this case can be excited by both TM and TE waves [3]. For example, optical bistability has been discovered recently in the transmission spectrum [4] and in the Goos–Hanchen shift [5] of such a nonlinear SP system. Moreover, in the literature, including some very recent studies on such nonlinear SP's [6–8], Eq. (1) has been inadvertently applied with the dielectric function  $\varepsilon_d$  simply be substituted by an intensity-dependent expression, e.g.,

$$\varepsilon_d = \varepsilon + \alpha |E|^2, \quad (2)$$

where  $\varepsilon$  and  $\alpha$  are the linear and nonlinear dielectric coefficients, respectively, and are frequency-dependent in general. However, since Eq. (1) is derived from Maxwell's equations by matching the appropriate boundary conditions for evanescent wave solutions with restriction only to dielectrics with a linear response, the above approach to the study of dispersion of the SP's at a metal–Kerr medium interface is not justified and can lead to appreciable errors in case of strong field intensity.

It is the purpose of this work to present an analysis of the magnitudes of these errors adopting such an approach to treat the nonlinear SP at a metal–Kerr medium interface. We shall apply a correct method treating this nonlinear SP's available from the literature and shall present numerical analysis on the limitation of the above inexact treatment by comparing it with results obtained from the correct theory.

## 2. Theory

We start with a brief recapitulation of the theory first formulated by Mihalache and coworkers. Following Ref. [9], we consider

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a TM-polarized wave at the interface of a Kerr-type medium and a metal, ignoring any loss in the media. Note that for simplicity we have assumed an isotropic Kerr medium as described in Eq. (2) for illustrative purpose. Generalization to the case of anisotropic media is rather straight-forward but mathematically tedious [9]. The electric field in both the nonlinear medium ( $z > 0$ ) and the metal ( $z < 0$ ) can be expressed as

$$\vec{E}(\vec{r}, t) = \frac{1}{2} \{iE_x(z)\hat{x} + E_z(z)\hat{z}\} \exp[i(\omega t - kz)] + c.c., \quad (3)$$

with the  $x$ - $z$  plane being the plane of incidence (Fig. 1), and it has been taken into account that the relative phase between the two components for the TM mode is  $\pi/2$ . This follows from Gauss's law together with our focus in looking for evanescent waves at the interface where propagation is permitted only along the  $\hat{x}$  direction [10]. The equations for the field amplitudes  $E_x(z)$ ,  $E_z(z)$  and  $H_y(z)$  in the Kerr medium were obtained from Maxwell equations in Ref. [9] in the form:

$$\frac{d}{dz} E_x = \frac{1}{k} \left( \frac{\omega^2}{c^2} \varepsilon_d - k^2 \right) E_x, \quad (4)$$

$$\frac{d}{dz} \varepsilon_d E_z = -k \varepsilon_d E_x, \quad (5)$$

$$\text{and } H_y = -\frac{\omega}{kc} \varepsilon_d E_z. \quad (6)$$

In the metal ( $z < 0$ ) which has a dielectric function  $\varepsilon_m$ , the  $x$ -component electric field amplitude will have the form  $E_x(z) = E_{0x} \exp(qz)$  where  $q^2 = k^2 - \frac{\omega^2}{c^2} \varepsilon_m$ . Thus  $\vec{\nabla} \cdot \vec{D} = 0$  implies

$$D_z = -\frac{k \varepsilon_m}{q} E_x. \quad (7)$$

The continuity of  $E_x$  and  $D_z$  across  $z = 0$  then yields

$$E_{0x} = -\frac{q}{k \varepsilon_m} (\varepsilon + \alpha E_0^2) E_{0z}, \quad (8)$$

with  $E_{0x} \equiv E_x(z = 0^+)$ ,  $E_{0z} \equiv E_z(z = 0^+)$  and  $E_0$  is the magnitude of the field. By using Eqs. (4) and (8) and a certain first integral obtained in Ref. [11], one can obtain another relation between the amplitudes  $E_{0x}$  and  $E_{0z}$  as follows [9,12]:

$$(\alpha/2) E_{0x}^4 + [\varepsilon + (\varepsilon_m \omega / cq)^2] E_{0x}^2 + (k \varepsilon_m E_{0z} / q) E_{0x} - (\alpha/2) E_{0z}^4 = 0. \quad (9)$$

Furthermore, using Eq. (8) together with the relation  $E_0^2 = E_{0x}^2 + E_{0z}^2$ , the following expressions can be derived:

$$E_{0z}^2 = \frac{\varepsilon_m^2 k^2 E_0^2}{(q^2 \alpha^2 E_0^4 + 2q^2 \varepsilon \alpha E_0^2 + q^2 \varepsilon^2 + k^2 \varepsilon_m^2)} = \frac{\varepsilon_m^2 k^2 E_0^2}{(q^2 \varepsilon_d^2 + k^2 \varepsilon_m^2)},$$

$$E_{0x}^2 = \frac{(\alpha^2 E_0^4 + 2\varepsilon \alpha E_0^2 + \varepsilon^2) q^2 E_0^2}{(q^2 \alpha^2 E_0^4 + 2q^2 \varepsilon \alpha E_0^2 + q^2 \varepsilon^2 + k^2 \varepsilon_m^2)} = \frac{\varepsilon_d^2 q^2 E_0^2}{(q^2 \varepsilon_d^2 + k^2 \varepsilon_m^2)}, \quad (10)$$

$$E_{0x} E_{0z} = -\frac{q}{k \varepsilon_m} (\varepsilon + \alpha E_0^2) E_{0z}^2 = -\frac{q}{k \varepsilon_m} \varepsilon_d E_{0z}^2,$$

where  $\varepsilon_d$  is as given in Eq. (2) with  $|E|^2 = E_0^2$ .

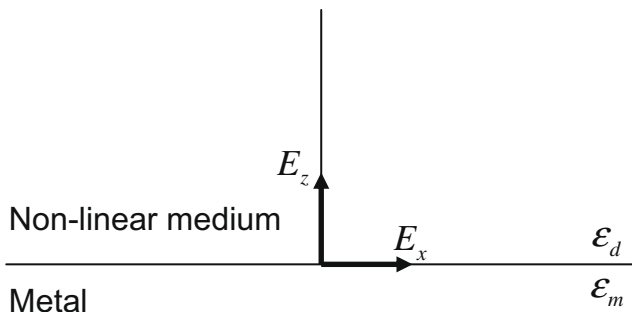


Fig. 1. Geometry of the problem.

Now let us apply the above results to derive an explicit and exact dispersion relation for the SP at a metal–Kerr dielectric interface. Substituting Eq. (10) into Eq. (9), the result can finally be reduced to a quadratic equation in the wave number  $k$  which, upon solving, yields the following relation:

$$k = \frac{\omega}{c} \frac{(\alpha E_0^2 + \varepsilon) \sqrt{\varepsilon_m} \sqrt{\alpha E_0^2 - 2\varepsilon_m + 2\varepsilon}}{\sqrt{\alpha^3 E_0^6 + 4\varepsilon \alpha^2 E_0^4 + 2\varepsilon^3 + 5\varepsilon^2 \alpha E_0^2 - 3\varepsilon_m^2 \alpha E_0^2 - 2\varepsilon_m^2 \varepsilon}} \quad (11)$$

$$= \frac{\omega}{c} \frac{\varepsilon_d \sqrt{\varepsilon_m} \sqrt{\varepsilon_d + \varepsilon - 2\varepsilon_m}}{\sqrt{(\varepsilon + \varepsilon_d) \varepsilon_d^2 - 3\varepsilon_m^2 \alpha E_0^2 - 2\varepsilon_m^2 \varepsilon}}$$

In the limit  $\alpha \rightarrow 0$  or  $E_0 \rightarrow 0$ ,  $\varepsilon_d \rightarrow \varepsilon$  and the result in Eq. (11) reduces back to Eq. (1) as expected. To find the surface plasmon resonance frequency  $\omega_{sp}$  from Eq. (11), one has to search for the zero's of the derivative  $d\omega/dk$ . However, this is equivalent to finding the poles of Eq. (11) since by writing (11) in the form:  $k = \omega f(\omega)$ , one obtains  $d\omega/dk = [f(\omega) + \omega f'(\omega)]^{-1}$ . Hence, the poles of  $f(\omega)$  is equivalent to the zeros of  $d\omega/dk$  since we expect the derivative  $f'(\omega)$  to be non-negative. Furthermore, one can show that the additional poles from  $f'(\omega)$  will be given by  $\varepsilon_m = 0$  and  $\varepsilon_m = (\varepsilon + \varepsilon_d)/2$ , which are unacceptable since these will make  $k = 0$  in Eq. (11). We thus obtain the following implicit expression for  $\omega_{sp}$  to satisfy from the poles of Eq. (11):

$$\varepsilon_m = -\sqrt{\frac{(\varepsilon + \varepsilon_d) \varepsilon_d^2}{\alpha E_0^2 + 2\varepsilon_d}}. \quad (12)$$

With the assumption of the ideal Drude model for the metal:

$$\varepsilon_m = 1 - \omega_p^2 / \omega^2, \quad (13)$$

the surface plasmon frequency can be solved from Eq. (12) to obtain:

$$\omega_{sp} = \omega_p / \left( \sqrt{1 + \left( \frac{(\varepsilon + \varepsilon_d) \varepsilon_d^2}{\alpha E_0^2 + 2\varepsilon_d} \right)^{1/2}} \right), \quad (14)$$

this result will be compared and contrasted with the one from the inexact approach [6–8], namely,

$$\omega_{sp} = \omega_p / \sqrt{1 + \varepsilon_d} = \omega_p / \sqrt{1 + \varepsilon + \alpha E_0^2}. \quad (15)$$

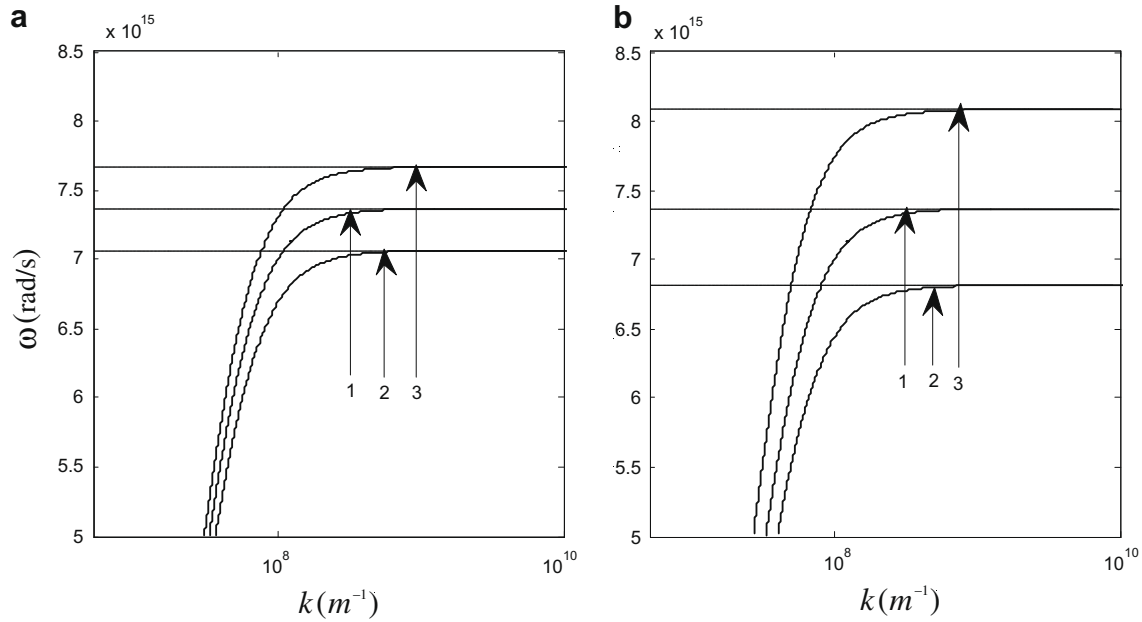
Note that even in the weak field limit, Eq. (14) implies

$$\omega_{sp} = \omega_p / \sqrt{1 + \varepsilon + (\alpha E_0^2 / 2)}, \quad (16)$$

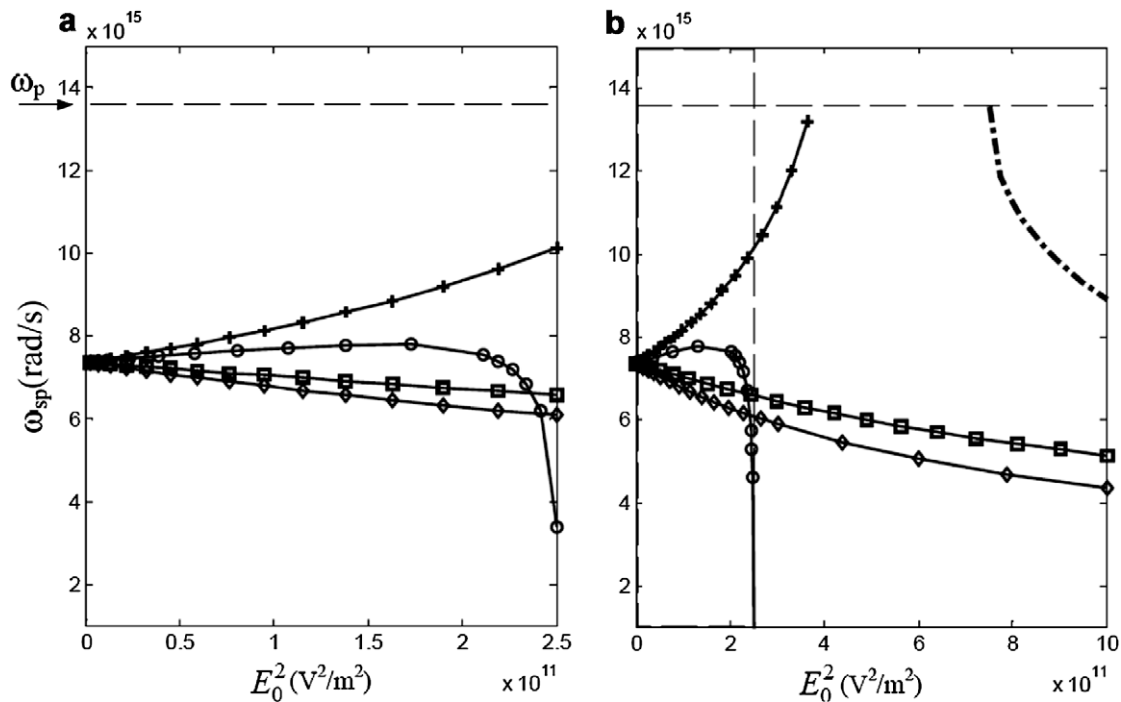
which contains an extra factor of 1/2 compared with the result in (15). In the following, we shall present numerical analysis of the inaccuracy in using Eq. (2) directly into Eq. (1) as done in some recent works [7,8] via comparison with exact results obtained from Eq. (11), for various magnitudes of the field intensity.

### 3. Numerical results and discussion

To evaluate the accuracy and limitation of the model used previously in the literature by combining Eqs. (1) and (2) [7,8], we have in Fig. 2 plotted the dispersion curves according to this model (Fig. 2b) and compared them with those from the exact model (Fig. 2a), i.e. Eq. (11). We have used the following values for the relevant parameters involved [9]:  $\alpha = \pm 6.4 \times 10^{-12} \text{ m}^2 \text{V}^{-2}$ ,  $\omega_p = 1.36 \times 10^{16} \text{ rad/s}$  (for Ag), and  $\varepsilon = 2.405$  for our computations, and we have ignored the dispersion of the coefficients for the Kerr medium. It is clear that the two agree only at zero electric field strength. Although both models show the correct trends for the dispersion curves to lie above ( $\alpha < 0$ ) and below ( $\alpha > 0$ ) the “linear



**Fig. 2.** Surface plasmon dispersion relations for a metal–Kerr interface plotted for different electric field strengths according to the two different approaches: (a) using consistent solutions from Maxwell’s equations, and (b) using result from linear optics. The nonlinear Kerr coefficient is fixed at  $|\alpha| = 6.4 \times 10^{-12} \text{ m}^2/\text{V}^2$  and the values of the field are as follows:  $E_0 = 0$  (curve 1) and  $E_0^2 = 9 \times 10^{10} \text{ V}^2/\text{m}^2$ , ( $\alpha > 0$ , curve 2) and ( $\alpha < 0$ , curve 3).



**Fig. 3.** Plot of the surface plasmon frequency as a function of the field intensity according to the two models described in Fig. 2: the consistent approach (' $\circ$ ' for  $\alpha < 0$  and ' $\square$ ' for  $\alpha > 0$ ); and the inexact model ('+' for  $\alpha < 0$  and '<' for  $\alpha > 0$ ). The value of  $\alpha$  is the same as that in Fig. 2, with Fig. 3a showing the portion of the results for smaller field values expanded i.e. the portion within the broken lines in (b). The dash-dotted line in (b) shows the “re-emerged solutions” for  $\alpha < 0$  as discussed in the text.

SP curve” (i.e.  $\alpha = 0$ ), they deviate appreciably from each other for larger values of  $E_0$  – with the exact model deviated from the “linear curve” by a less amount compared to the model from the approach using Eqs. (1) and (2). All these can easily be understood by simply referring to the low field limit of each of the exact model by noticing the extra  $\frac{1}{2}$  factor in Eq. (16) as pointed out above. The physical reason is that in the linear approximation model, the Kerr medium

is taken to behave as a homogeneous medium with a dielectric constant given by Eq. (2); whereas in the exact approach, the gradual decrease in the nonlinear term away from the interface is taken into account (due to the decay of the evanescent field away from the interface). This then leads effectively to a smaller change from the linear dielectric constant for the Kerr medium and hence a less amount of deviation from the “linear curve”. Fig. 3 shows the

comparison between the surface plasmon frequencies obtained from each of the two models, i.e. Eqs. (14) and (15), as a function of the field intensity. Again, it shows clearly that the inexact approach can yield incorrect values for the surface plasmon frequencies (both for  $\alpha < 0$  and  $\alpha > 0$ ) even for modest values of field strength. For example, at  $E_0 = 3.0 \times 10^5$  V/m which corresponds to a nonlinearly of about 19% in the Kerr dielectric function (and about 10% in the refractive index for the  $\alpha > 0$  case), the wrong approach has underestimated the surface plasmon resonance frequency by about 3.5% for positive nonlinearity ( $\alpha > 0$ ) and overestimated it by about 5.4% for  $\alpha < 0$ . Note that this latter case is of great interest since the correct approach predicts a complete opposite trend for further increase of field strength. With the limitation in the magnitude of the field to guarantee  $\omega_{sp} < \omega_p$ , one sees that as the field increases, the inexact approach predicts  $\omega_{sp} \rightarrow \omega_p$  monotonically while the exact approach predicts a much slower rise and then a sudden decrease in the resonance frequency which can become exceedingly small corresponding to the condition  $\alpha E_0^2 + 2\varepsilon_d \approx 0$  in Eq. (14). This sharp difference between the two models for  $\alpha < 0$  should be testable relatively easy in an experiment. For further increase of the field intensity, the exact approach predicts that in the  $\alpha < 0$  case, there is no solution for  $\omega_{sp}$  for the intensity range  $\frac{2\varepsilon_c}{3|\alpha|} < E_0^2 < \frac{2\varepsilon_c}{|\alpha|}$  but solution (shown as dash-dotted line) re-emerges for larger values of  $E_0$  (i.e. for  $E_0^2 > 7.7 \times 10^{11}$  V<sup>2</sup>/m<sup>2</sup>). However, these re-emerged solutions are likely not of much physical significance, since for these large field values, we will have  $\varepsilon_d < 0$  which implies a negative SP wave number ( $k$ ) from Eq. (11). Furthermore, for the range of  $E_0$  corresponding to the re-emerged solution, we have tested numerically Eq. (11) and found that it leads to imaginary values for  $k$ . Hence, it is more likely that for the  $\alpha < 0$  case, the correct treatment also yields a kind of “cut-off” field intensity value (at  $E_0^2 = \frac{2\varepsilon_c}{3|\alpha|}$ ) similar to that exists in the linear model, except the trend of the resonance frequency is completely opposite as this value of field intensity is approached. As for the other features in Fig. 3, most of them can easily be understood if we consider the expression  $(\frac{\varepsilon_c + \varepsilon_d + \alpha E_0^2}{\alpha E_0^2 + 2\varepsilon_d})^{1/2}$  as an “effective dielectric function” for the Kerr medium as implied from Eq. (14).

#### 4. Conclusion

In this communication, we have analyzed the inaccuracy of a certain approach to the study of the surface plasmon excitations at a Kerr–metal interface as implemented in some recent works [7,8] adopting results from linear optics. For simplicity we have limited ourselves to isotropic and lossless media. The generalization to anisotropic Kerr media is possible with some mathematical complications [9], but the inclusion of loss in such a nonlinear optical problem will be more challenging and will be left for future investigations. However, from the results for the case of linear SPR, it is known that dissipation in the metal will lead to a similar dispersion relation as in Eq. (1), with only the real part of the metallic dielectric function entering the equation provided that the loss is small [1]. Though we expect the same will likely happen

to our results in Eqs. (11) and (14), it will be of interest to clarify this in a rigorous way. In addition, the finite range of propagation of the SP along the interface can only be obtained by accounting for the loss in the metal [1].

In the present work, we have emphasized that a correct approach must start afresh from the Maxwell wave equation [9] which can yield very different results even for modest values of the electric field strength. The discrepancy between the two approaches is especially significant for the case with a negative nonlinear term in the Kerr dielectric function. It would be of great interest if the drastically different results predicted by the correct model for the dispersion relations in this case can be verified experimentally in a simple way, such as that from the angular spectrum of a Kretschmann ATR experiment [1], as was employed previously in the observation of bistability in the reflection spectrum of a metal–Kerr medium system [13].

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