Numerical study of surface plasmon enhanced nonlinear absorption and refraction

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Abstract: Maxwell Garnett effective medium theory is used to study the influence of silver nanoparticle induced field enhancement on the nonlinear response of a Kerr-type nonlinear host. We show that the composite nonlinear absorption coefficient, β_c, can be enhanced relative to the host nonlinear absorption coefficient near the surface plasmon resonance of silver nanoparticles. This enhancement is not due to a resonant enhancement of the host nonlinear absorption, but rather due to a phase-shifted enhancement of the host nonlinear refractive response. The enhancement occurs at the expense of introducing linear absorption, α_c, which leads to an overall reduced figure of merit β_c/α_c for nonlinear absorption. For thin (< 1 μm) composites, the use of surface plasmons is found to result in an increased nonlinear absorption response compared to that of the host material.

OCIS codes: (260.2065) Effective medium theory; (160.1245) Artificially engineered materials; (160.4330) Nonlinear optical materials; (160.4236) Nanomaterials

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1. Introduction

The fabrication of materials that exhibit strong nonlinear absorptive properties at visible and near-infrared frequencies has been of interest for several years [1-8]. Such materials would enable control over the maximum transmitted irradiance or fluence through a material, which could be used for example to minimize optically induced damage while still allowing non-damaging levels of radiation to pass through [3, 5]. One approach to achieve this involves the use of materials that exhibit relatively large two-photon absorption (TPA). Due to the small values of the nonlinear absorption coefficient in most materials, the nonlinear absorption that can be achieved in this manner is generally weak. A common approach to increase the nonlinear absorption is to use lenses that focus the incident light onto the nonlinearly absorbing sample [3, 5], resulting in a larger irradiance and therefore inducing a larger nonlinear absorption. A significant drawback of this approach is the relatively complicated and bulky nature of the resulting optical system. To simplify such systems it would be beneficial to design materials with a strongly enhanced nonlinear optical (NLO) response, potentially removing the need for external lenses.

An alternative approach to achieving an enhanced nonlinear response involves the use surface plasmon resonances. A surface plasmon (SP) is a collective oscillation of free charges in a conductor that, when resonantly excited, can induce strongly enhanced local fields. For example, isolated silver nanoparticles in water can produce local field enhancements at optical frequencies in the range of 10 - 40. As we will show, this field enhancement can significantly increase the magnitude of the nonlinear refractive index. In order to evaluate the effect of SP-induced field enhancement on the nonlinear absorption of a host material, we investigate the

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characteristics of a composite consisting of a small volume filling fraction of metal nanoparticles (NPs) in a nonlinear host material. The NLO properties of such dilute metallodielectric composites have been studied extensively both theoretically [9-12] and experimentally [13-25]. Many of the experiments reported in literature consider the nonlinear response at a single wavelength or discuss the influence of surface plasmons on the nonlinear response of the composite material while ignoring the effect of the associated changes to the linear response on the figure of merit β_c/α_c where β_c is the effective composite nonlinear absorption coefficient and α_c is the composite linear absorption coefficient.

In the present theoretical study we investigate the nonlinear optical properties of a metal dielectric composite over a broad range of frequencies and explicitly evaluate the effect of surface plasmon-induced linear absorption on the nonlinear absorptive performance. In particular we consider the effect of surface plasmons on the nonlinear response of the *host material*, ignoring any nonlinearity of the metal itself. We analytically calculate the linear optical response of the composite material and evaluate its effect on the maximum nonlinear absorption that can be achieved in a composite material of finite thickness. Using a realistic model system consisting of metal nanoparticles embedded in a realistic highly nonlinear host material, we show that the SP mediated field enhancement in a nonlinear *refractive* host results in a large positive nonlinear *absorption*. Furthermore we show that for very thin samples in which the SP induced linear absorption is relatively low, the total nonlinear absorption of the composite can be improved significantly by the addition of metal nanoparticles. Finally, we show that the presence of a strong SP related linear absorption in these composites puts a practical upper bound on the thickness of the surface plasmon-enhanced nonlinear absorbing composite.

2. Computation of effective medium parameters

2.1. Model system

In order to study the effect of surface plasmons on the nonlinear absorption of a composite material, we investigate a model system that is expected to provide a large and fast nonlinear response. The host material is chosen to be carbon disulfide (CS_2) which is a material with a large intrinsic nonlinear absorption cross section at the surface plasmon resonance for Ag. The linear properties for CS_2 are taken from [26] and the complex nonlinear refractive index of CS_2 is taken to be $n_{2h}=6\times10^{-15}$ cm^2/W with β_h=0.6 cm/GW, as obtained by z-scan measurements using low repetition rate fs pulses at a center wavelength of 440 nm [27]. In the present study these values are assumed to be constant at wavelengths close the nanoparticle resonance (380-500nm).

To enhance the nonlinear response of CS_2, we consider 10 nm diameter spherical silver nanoparticles, which are well known to support surface plasmon resonances in the ultraviolet to blue spectral region with large local field correction factors. For the description of the silver nanoparticles in the frequency range of interest we use a surface scattering corrected Drude model fit to literature values obtained by Johnson and Christy [28]. The complex dielectric function of the silver inclusions ε_i as a function of angular frequency ω [rad/s] is given by

$$\varepsilon_i = \varepsilon_\infty - \omega_p^2 \Big/ \left[\omega^2 + i(\Gamma_0 + \Gamma_s)\omega\right] \tag{1}$$

using fitting values of ε_∞=5.451, $\omega_p=1.474\times10^{16}$ [rad/s], and $\Gamma_0 = 8.354\times10^{13}$ [s^{-1}], where Γ_0 is the fitted bulk electron scattering rate. The estimated surface scattering rate is given by $\Gamma_s = A\upsilon_f/r = 2.8\times10^{14}$, where A is a constant set to 1, $\upsilon_f = 1.39\times10^6$ m/s is the Fermi velocity in silver, and r is the radius of the particle. As will be shown below, the enhancement of the nonlinear absorption coefficient depends strongly on the exact values used for the complex dielectric function of the metal. This makes the consideration of surface scattering essential when calculating the nonlinear response of silver nanoparticle-based composites. For example, a nanoparticle diameter as large as 350 nm is sufficient to change the total damping rate by 10% relative to the measured bulk values.

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2.2 Linear effective medium properties

The linear optical properties of a system consisting of a low volume fraction of non-interacting small spherical particles embedded in a host material are well described by Maxwell Garnett (MG) theory [9, 10], under certain limiting conditions. The first condition is that the optical response of the system can be well approximated by the electrostatic limit. This requires the nanoparticles to be much smaller than the incident wavelength in the host material *as well as* smaller than the skin depth. Full field simulations of Ag particles in CS_2 under plane wave illumination (not shown) indicate that both requirements are satisfied for 10 nm diameter silver particles. The second assumption is that neighboring particles do not interact. This requirement can be satisfied by working with low volume filling fractions. In this study we have assumed a volume filling fraction, f, of 10^{-3} unless otherwise stated. For 10 nm diameter particles, this corresponds to a typical center-to-center particle spacing of the order of 100 nm. Under these conditions, the dielectric function of a MG composite material, ε_c, is given by

$$\varepsilon_c = \left(\frac{1+2f\gamma}{1-f\gamma}\right)\varepsilon_h \quad \gamma = \frac{\varepsilon_i - \varepsilon_h}{\varepsilon_i + 2\varepsilon_h} \tag{2}$$

where ε_i is the dielectric function of the inclusion (here Ag), and ε_h is the dielectric function of the host material (here CS_2). The parameter γ is a field enhancement factor representing the ratio of the internal particle response field, resulting from induced surface charge, relative to the incident field.

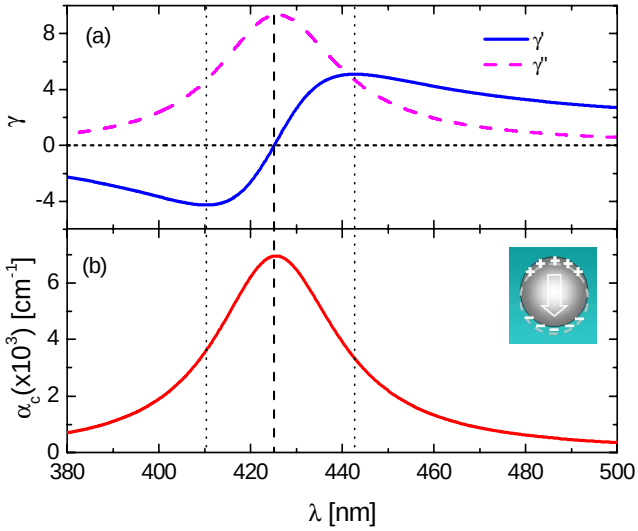

Fig. 1. (a) Real and imaginary parts of the local field correction factor and (b) composite linear absorption.

Figure 1(a) shows a plot of the real and imaginary parts of γ obtained from Eq. (2). A resonance feature is observed at a wavelength of 425 nm. This resonance coincides with the frequency at which the denominator of the expression for γ reaches a minimum, which occurs when $\varepsilon_i' = -2\varepsilon_h'$ with ε' indicating the real part of the respective dielectric functions. This feature is due to the excitation of the Fröhlich mode or the nanoparticle plasmon resonance, in which the electrons oscillate in a dipolar manner, as shown in the inset in Fig. 1(b). In this and further plots, the dashed vertical line indicates the nanoparticle plasmon resonance frequency, and the dotted vertical lines represent the half width at half maximum points of the plasmon resonance, or more precisely, of the imaginary part of the field enhancement factor,

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γ. Note that on resonance, γ is almost entirely imaginary, indicating that the response field inside and outside the particle exhibit close to a 90° phase difference relative to the incident light (a 90° phase lead inside the particle, and a 90° phase delay outside the particle along the direction of polarization). This complex nature of the particle response has an important consequence: even if ε_h has a negligible imaginary component, the dielectric function of the composite, ε_c, will contain a nonzero imaginary part, resulting in a finite absorption coefficient. In Fig. 1(b) we show the resulting composite linear absorption coefficient, α_c. Near resonance, α_c reaches a peak value of ~7000 cm^{-1}. This shows one major challenge in the use of surface plasmons to enhance nonlinear absorption: the presence of resonantly enhanced linear absorption limits the useful sample thickness. This will be discussed in more detail below.

2.3 Nonlinear effective medium properties

The enhanced fields that occur near metal nanoparticles as a result of the surface plasmon resonance give rise to an enhanced nonlinear polarization of the host. The MG theory has been extended to compute the resulting nonlinear susceptibility enhancement factor $g^{(3)}$ of the third order susceptibility of a metal-dielectric composite assuming that the metal response is entirely linear, and that the host exhibits a nonlinear refractive index [9, 10]. The resulting third order susceptibility enhancement factor is given by

$$g^{(3)} \equiv \frac{\chi_c^{(3)}}{\chi_h^{(3)}} = \frac{1}{5}\left|\frac{\varepsilon_c + 2\varepsilon_h}{3\varepsilon_h}\right|^2 \left(\frac{\varepsilon_c + 2\varepsilon_h}{3\varepsilon_h}\right)^2 (1-f)[8f(1+f+f^2)|\gamma|^2\gamma^2 + 6f(1+f)|\gamma|^2\gamma + 2f(1+f)\gamma^3 + 18f(|\gamma|^2+\gamma^2) + 5] \quad (3)$$

where γ is defined in Eq. (2). At low filling fractions (i.e. f«1 and $\varepsilon_c \approx \varepsilon_h$) this equation simplifies to

$$g^{(3)} \equiv \frac{\chi_c^{(3)}}{\chi_h^{(3)}} = 1 + \frac{f}{5}\left[8|\gamma|^2\gamma^2 + 6|\gamma|^2\gamma + 2\gamma^3 + 18\left(|\gamma|^2+\gamma^2\right)\right]. \quad (4)$$

The total enhancement of the third order susceptibility is seen to depend on the field enhancement factor according to terms of magnitude $|\gamma|^4$, $|\gamma|^3$, and $|\gamma|^2$. This complicated form is a consequence of the inhomogeneous distribution of the electric field outside the polarized nanoparticle. Note that the maximum value of γ occurs near resonance and scales with $1/\varepsilon_i$". As a result, the magnitude of the nonlinear enhancement factor depends strongly on the imaginary part of the dielectric function. Although the trends observed in this study are general, the results will depend on the exact values of imaginary part of the dielectric function of the metal and of the host.

In order to evaluate the NLO refractive properties of the composite material, we calculate the complex nonlinear refractive index $\eta_2 = n_2 + i\kappa_2$ based on the composite nonlinear susceptibility $\chi_c^{(3)} = g^{(3)}\,\chi_h^{(3)}$ as obtained from Eq. (3) and using the known dielectric functions. Since our composite medium has a complex linear refractive index, we use the general equation relating the third order susceptibility to the nonlinear complex refractive index, which is given in SI units by [29]

$$\eta_2\left[\frac{m^2}{W}\right] = \frac{3}{4\varepsilon_0 c\,|\eta|^2}\left(1 - i\frac{\kappa}{n}\right)\chi^{(3)} \quad (5)$$

where n and κ are the real and imaginary parts of the linear refractive index respectively and $\chi^{(3)}$ is the nonlinear susceptibility in units of m^2/V^2. We can now define a *nonlinear refractive index enhancement factor*, g_2, which is given by

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$$g_2 = \frac{\eta_{2,c}}{\eta_{2,h}} = \frac{|\eta_h|^2}{|\eta_c|^2} \frac{1 - i(\kappa_c / n_c)}{1 - i(\kappa_h / n_h)} g^{(3)} \tag{6}$$

Note that with the exception of extremely lossy composites, the frequency dependent nonlinear index enhancement factor, g_2, will be virtually identical to the third order susceptibility enhancement factor, $g^{(3)}$.

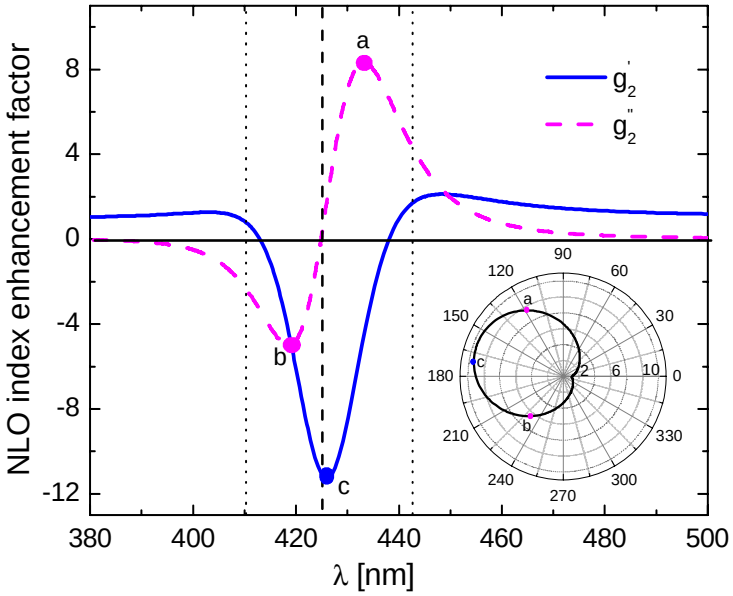

Fig. 2. Real and imaginary parts of the nonlinear refractive index enhancement factor g_2 of 10 nm diameter Ag particles embedded in CS_2.

Figure 2 shows the complex nonlinear index enhancement factor, g_2, for our composite as obtained using Eq. ((6). Near resonance, the nonlinear refractive index enhancement factor is a complex quantity, which is a consequence of the previously discussed frequency dependent phase shift between the incident field and the local field. The inset shows the same data displayed in terms of complex phase and amplitude, with increasing wavelength corresponding to clockwise rotation along the curve. These results can be used to predict the influence of the surface plasmon resonance on the response of a nonlinear host material. Since g_2' is negative over most of the plasmon resonance, the use of a host material with a large positive κ_{2h} (and thus a large effective nonlinear absorption coefficient) will in fact result in a composite that has a large negative κ_{2c} corresponding to a high effective *saturable absorption* at the surface plasmon resonance frequency. These results demonstrate that surface plasmons do not directly enhance the nonlinear absorption coefficient of a host material, but rather provide a phase-shifted enhancement of the complex nonlinear refractive properties of the host. As a result, enhanced nonlinear absorption can be achieved in a narrow frequency range (a) *below* the plasmon resonance by using a positive nonlinear refractive host, (b) *above* the plasmon resonance by using a negative nonlinear refractive host, or, in principle, (c) *at* the plasmon resonance by using a saturable absorbing host. To our knowledge, however, an instantaneous, homogeneous saturable absorbing host does not exist.

Figure 3 shows the calculated composite nonlinear refractive index, n_{2c}, (solid line, panel (a)) and the nonlinear absorption β_c (solid line, panel (b)) as a function of wavelength for Ag nanoparticles in CS_2. As we stated in section 2.1, these and subsequent calculations assume

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that the values of n_{2h} and β_h do not change over the wavelength range of interest. At the SP resonance frequency the composite exhibits strongly negative nonlinear refraction, despite the positive nonlinear refractive properties of the host. At frequencies slightly below the SP resonance the composite can be seen to exhibit strong nonlinear absorption, with a β_c that is ~20 times larger than β_h. This is an impressive result, given the small metal fill fraction of $f = 10^{-3}$. As discussed above, it is important to realize that this large β_c value is *not* due to a resonant enhancement of β_h, but rather due to a *phase-shifted enhancement of the nonlinear refractive response of the host*. To highlight this, the dotted lines in Fig. 3 show the nonlinear refractive properties of the composite that result exclusively from the enhancement of β_h, while the dashed line shows the nonlinear refractive properties of the composite that result exclusively from the enhancement of $n_{2,h}$. From Fig. 3 it is clear that the composite nonlinear response is dominated by the enhanced nonlinear refractive response of the host material for most frequencies around the SP resonance. Note that the frequency bandwidth over which enhanced β values are obtained is less than half the SP resonance width, due to the more than quadratic dependence of the nonlinear index enhancement on the field enhancement factor.

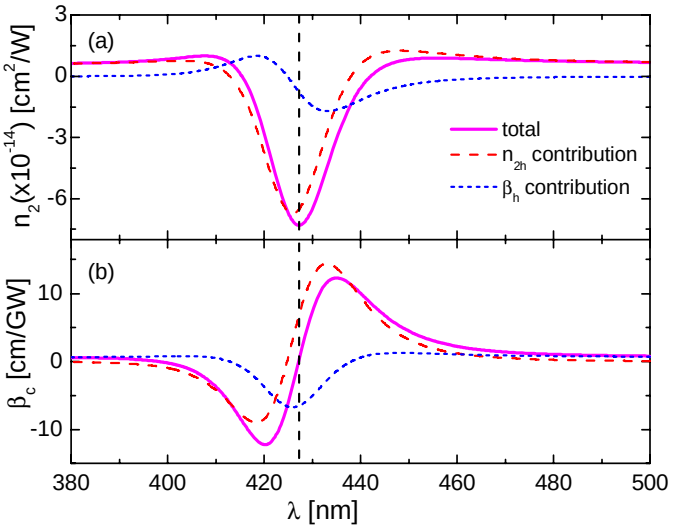

Fig. 3. (a) Calculated nonlinear refractive index and (b) nonlinear absorption coefficient of a Ag-CS_2 nanocomposite. The separate contributions from the host nonlinear refractive index and host nonlinear absorption are indicated by the dashed and dotted curves respectively.

The appearance of nonlinear absorption in a composite material consisting of *linear* nanoparticles and a nonlinear *refractive* host is counterintuitive, but can be understood by considering the effect of the host refractive index on the plasmon resonance frequency. From Eq. (2) it can easily be seen that the surface plasmon resonance shifts to lower frequencies as the refractive index of the host is increased, taking into account that the metal dielectric function becomes more negative at lower frequencies. For a nonlinear host with a positive value of n_2, a high irradiance is accompanied by an increase in the refractive index of the host, resulting in a redshift of the plasmon resonance. This simple observation can qualitatively explain the appearance of nonlinear absorption. To illustrate this point, Fig. 4(a) shows the calculated linear absorption spectrum (solid black line) of an Ag-CS_2 composite at a fill fraction of 10^{-3}, as well as the corresponding absorption spectrum when the refractive index of the host is artificially increased by 0.01 (gray solid line). To improve the visibility of the small resulting changes in the absorption spectrum, the values ε_{Ag}'' are reduced by a factor ten and a large index change of 0.01 is used. The absorption spectrum clearly exhibits a redshift as a result of the refractive index increase. This shift can be seen to lead to a decrease in absorption at high frequencies (downward arrow), corresponding to apparent saturable

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absorption. Similarly, at low frequencies this same shift leads to an increase in absorption (upward arrow), corresponding to nonlinear absorption. These observations are in agreement with the trends observed in the calculated composite nonlinear absorption resulting from n_{2h}, as shown in Fig. 4(b).

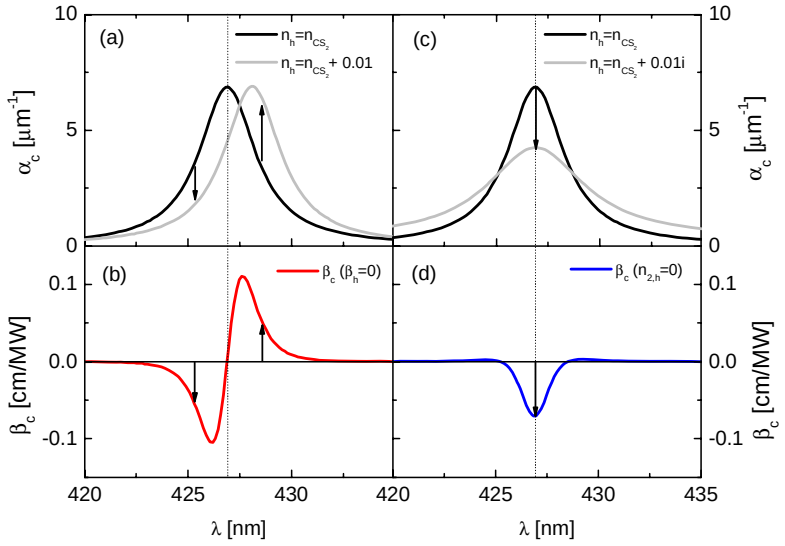

Fig. 4. (a) Calculated linear absorption of a Ag-CS_2 composite with and without a small increase in the host refractive index. (b) Calculated composite nonlinear absorption coefficient considering only the nonlinear refractive index contribution of the host. (c) Calculated linear absorption of a Ag-CS_2 composite with and without a small increase in the absorption coefficient of the host. (d) Calculated composite nonlinear absorption coefficient considering only the nonlinear absorption contribution of the host.

Similarly, the effect of a nonlinear absorptive host on the composite nonlinear response can be understood by considering its effect on the surface plasmon linewidth. For a host with a positive value of β, a high irradiance is accompanied by an increase in the host absorption. From Eq. (2), it can be seen that the introduction of an imaginary contribution to the host dielectric function results in a broader and weaker surface plasmon related absorption response in the composite. To illustrate this, Fig. 4(c) again shows the linear absorption calculated for a Ag-CS_2 composite (solid black line), as well as for a composite where the imaginary part of the host refractive index has been artificially increased by 0.01 (solid grey line). From Fig. 4(c) we see that the resulting broadening of the plasmon resonance leads to a decrease in absorption near the center of the resonance, corresponding to an effective saturable absorption. At frequencies away from the surface plasmon resonance, a slight increase in absorption is observed, corresponding to nonlinear absorption. These observations are in agreement with the trends observed in the calculated composite nonlinear absorption resulting from β_h, shown for comparison in Fig. 4(d). Similar arguments may be used to understand the effect of a nonlinear inclusion on the composite nonlinear response. Note that this analysis ignores the inhomogeneous nature of the electric field throughout the composite, and as such only provides a qualitative understanding of the processes involved. Fig. 4(a) does point out an important effect: at the high index changes shown, the nonlinear absorption is clearly no longer linearly dependent on the incident irradiance. This suggests that considering only third order nonlinearities is only accurate when the induced plasmon resonance frequency shift is much smaller than the plasmon resonance linewidth.

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3. Evaluation of plasmon enhanced nonlinear absorption

3.1 Figure of merit (FOM)

In order to evaluate the nonlinear optical response of different MG composites, in Fig. 5(a) we show the computed β_c for fill fractions, *f*, ranging from 10^{-6} to 10^{-3}. At low filling fractions the composite nonlinear absorption closely resembles that of the host. As the fill fraction is increased, the nonlinear absorption coefficient of the composite is seen to change significantly at frequencies near the plasmon resonance. For this composite, these changes become significant only at fill fractions larger than ~10^{-4}. The peak enhancement of β_c continues to increase as the fill fraction is raised. This suggests that the strongest nonlinear optical absorption will be achieved at the largest possible fill fraction. However as discussed in the introduction, the presence of an SP resonance also results in enhanced linear absorption, putting an upper bound on the maximum practical thickness of the composite. For this reason a figure of merit (FOM) is commonly employed defined as FOM = β_c/α_c in order to reflect the fact that the maximum useful thickness z_{max} is approximately $1/\alpha_c$.

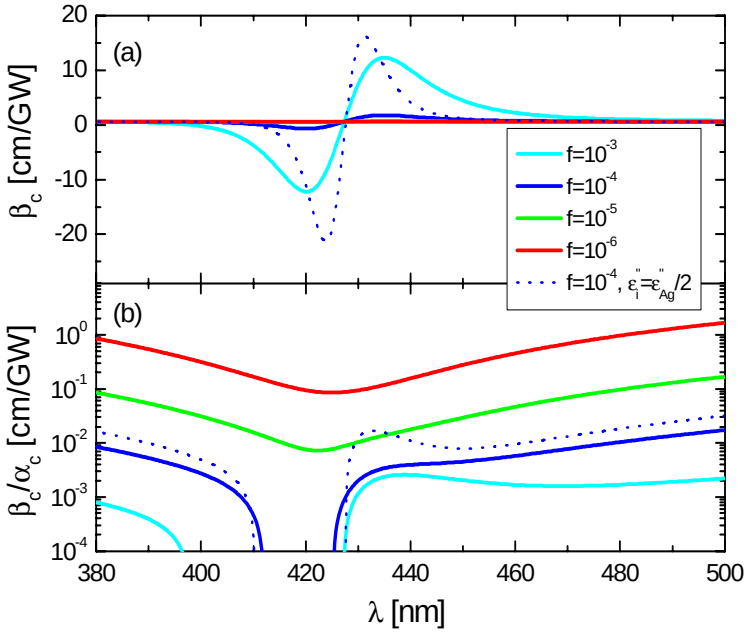

Fig. 5. (a) Nonlinear absorption coefficient β_c of a silver-CS_2 composite for several silver filling fractions and (b) corresponding figure of merit.

Figure 5(b) shows the calculated frequency dependent FOM of a CS_2-Ag nanoparticle composite material for several filling fractions based on the calculated β_c and the linear absorption coefficient α_c calculated using Eq. (2). Perhaps surprisingly, despite the observed strong enhancements in β and n_2, the highest figure of merit is observed for the lowest metal fill fraction and away from the SP resonance. This shows that if the use of thick samples is acceptable, it is preferable to avoid the use of surface plasmon mediated field enhancement, since in this case the benefits of enhancing the nonlinear optical response are outweighed by the introduction of strong linear loss. We will discuss this in more detail in the next section. Note that a negative FOM is obtained at frequencies just above the SP resonance for the 10^{-4} and 10^{-3} filling fractions, corresponding to an induced effective saturable absorption, rather than the desired enhanced nonlinear absorption. It should be pointed out that for a given host, an independent method to increase the peak β_c is to reduce the loss in the metal (Eqs. (3) and (6)). This increase in β_c would be accompanied by a corresponding reduction in the resonance bandwidth and an increase in the peak α_c (Eq. (2)). To illustrate this effect, the dotted lines in

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Fig. 5 show the frequency dependent β_c and FOM at a fill fraction f=10^{-4} when ε_i'' is reduced by a factor of 2. Figure 5(a) demonstrates that this reduction in loss leads to an increase in the peak value of β_c of close to an order of magnitude. In addition, from Fig. 5(b), it can be seen that the reduction in loss leads to an overall improvement of the figure of merit, indicating that the increase in linear absorption is outweighed by the increase in nonlinear absorption.

3.2 Nonlinear transmission

In order to exemplify the interplay between the composite linear and nonlinear absorption, we calculate the transmitted irradiance (I_{out}) as a function of incident irradiance (I_{in}) at a wavelength of 435 nm. This corresponds to the wavelength at which β_c is approximately maximum for all fill fractions (see Fig. 5(a)), and as such the curves represent a 'best case scenario'. In the presence of both linear and nonlinear absorption, the transmitted irradiance is given by

$$I_{out} = \frac{-\alpha_c I_{in}}{\beta_c I_{in} - (\alpha_c - \beta_c I_{in}) e^{\alpha_c L}} \tag{7}$$

where L is the sample thickness. Note that this equation assumes normal incidence plane wave illumination.

Figure 6(a) shows the nonlinear transmission for a 100 nm thick composite Ag-CS_2 layer. This thickness is sufficiently small to prevent any significant linear absorption for the fill fractions used, as evidenced by the fact that at low incident irradiance the transmitted irradiance is independent of fill fraction. For reference the linear transmission corresponding to zero absorption is indicated by the dashed line. At higher irradiances we see that I_{out} begins to deviate from linear transmission as a result of the finite β_c. The point at which the transmitted irradiance deviates from a linear response by 50% is indicated by the solid circles. As the irradiance increases further, the total transmitted irradiance is seen to saturate. A similar trend is observed for all fill fractions, however as the fill fraction is increased, the transmitted irradiance is seen to saturate at lower input irradiance, and the maximum transmitted irradiance is reduced. Both these effects are due to the fact that β_c increases as f increases, as was observed in Fig. 5(a). These results show that for thin layers, the nonlinear absorption can be enhanced by the addition of metal nanoparticles, without significantly affecting the linear transmission.

Figure 6(b) shows the calculated nonlinear transmission for a 10 μm thick composite Ag-CS_2 layer. Two important differences are observed compared to the thin layer results shown in Fig. 6(a). First, for small fill fractions (top curves) we see that a lower saturation irradiance is achieved than in Fig. 6(a) due to the longer interaction length. For low metal fill fractions, the nonlinear transmission is largely due to the response of the host material, with a negligible contribution from the small amount of metal in the composite. As the fill fraction is increased to 10^{-3}, the onset of nonlinear behavior is seen to shift to lower irradiance, as was observed in Fig. 6(a). In this case however, the linear transmission of these layers is also found to decrease dramatically as a result of the surface plasmon induced linear absorption. These results highlight the main challenge in the use of surface plasmons for the enhancement of nonlinear refractive and absorptive applications: although the nonlinear response can be significantly enhanced by the plasmon resonance, the induced linear loss places an upper bound on the useful sample thickness for a fixed metal fill fraction.

It should be pointed out that the irradiances assumed here in order to observe nonlinear transmission in very thin (<10 μm) layers exceed 1000 GW/cm^2. At these extremely high irradiance values nonlinear processes other than Kerr nonlinearities are expected to dominate, including dramatic thermal effects and possibly ionization and break down. It should therefore be emphasized that these curves indicate the limits of what can be achieved in such dilute systems *purely* based on a Kerr response in the host material. To experimentally observe these effects, thicker layers or higher fill fractions are likely needed, and special care

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will have to be taken to minimize thermal effects, including the use of femtosecond radiation at low repetition rates.

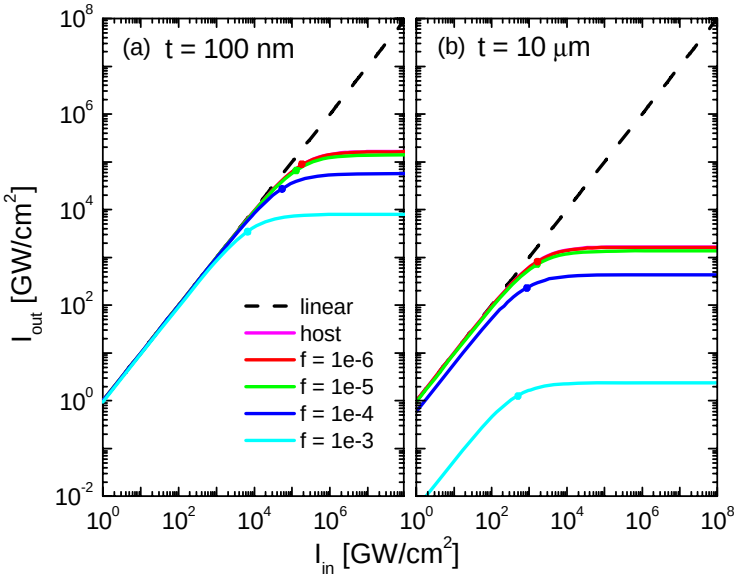

Fig. 6. Transmitted irradiance as a function of incident irradiance at a wavelength of 435 nm for different filling fractions for an Ag-CS_2 composite with a thickness of (a) 100nm (thin layer limit) and (b) 10 μm. The dashed line represents a linear response with zero absorption.

The results presented show that surface plasmons on metal particles can be used to strongly modify the nonlinear refractive properties of dilute metallodielectric composites. Based on our findings it is clear that the development of such systems for real applications should focus on the minimization of plasmon enhanced linear absorption while maintaining a significant enhancement of the nonlinear refractive response. Future work will focus on evaluating the response of composites that are outside the applicability of the Maxwell Garnett theory, such as dense structures containing non-spherical elements. Such dense plasmon enhanced nonlinear absorbing media could provide an improved balance between linear and nonlinear absorption.

4. Conclusion

The influence of plasmon resonant silver nanoparticles on the linear and nonlinear effective medium properties of CS_2 in the Kerr regime is explored for a Maxwell Garnett type composite. We demonstrate that the nonlinear absorption coefficient of a Ag-CS_2 composite can be enhanced relative to the nonlinear absorption coefficient of CS_2 over a narrow spectral range near the surface plasmon resonance. It is shown that this enhancement is not due to the enhancement of the host nonlinear absorption coefficient but rather due to a phase shifted enhancement of the host nonlinear refractive index. Furthermore it is shown that this effect occurs at the expense of the introduction of a surface plasmon related linear absorption to the composite resulting in an overall reduced FOM for composites containing metal nanoparticles. The induced linear absorption has the effect of placing an upper limit on the useful sample thickness. For a volume filling fraction of 10^{-3} this results in an upper useful sample thickness of ~1 μm at the wavelength of maximum linear absorption. For thin samples (linear transmission > 1/e) the total nonlinear absorption of the composite can be enhanced relative to that of the host alone at the same thickness. However, due to the dependence of the total nonlinear absorption on the sample thickness, a stronger nonlinear absorption can be obtained by avoiding the use of silver nanoparticle plasmon resonances and making use of a thicker layer. For a given host, reducing the imaginary part of the metal dielectric function

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results in a stronger but narrower resonance, and an improved ratio between the nonlinear and linear absorption coefficient.

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