Study of Non-linear Optical Properties of an ENZ Composite Metamaterial

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We hereby certify that the project which is presented in the Major Project – II entitled "Study of Non-linear Optical Properties of an ENZ Composite Metamaterial" and submitted to the Department of Applied Physics, Delhi Technological University, Delhi is record of our own, carried out during the period of January to May 2022, under the supervision of Prof. Ravindra Kumar Sinha.

The matter presented in this report has been submitted by us for the award of this degree any other degree of this or any other Institute/University. The work has been communicated in peer reviewed Scopus indexed conference with the following details:

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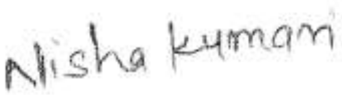

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ABSTRACT

We designed a non-linear composite metamaterial made up of alternating layers of fused silica glass and silver with thicknesses adjusted so that ε'=0 in the near infrared range., and the structure's optical behaviour is theoretically studied. It shows a shift from negative to positive in the real component of dielectric permittivity as a function of wavelength. we also have studied the variation of ENZ wavelength with thickness of dielectric layer and plasma frequency. By using the Maxwell Garnett theory, we have calculated non-linear optical properties, and we find that as we increase the thickness of dielectric material the ENZ wavelength increases along with the increases in real part of non-linear susceptibility. A significant enhancement in nonlinear parameters has been also observed at ENZ wavelength due to the ENZ nature of the composite material.

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List of Symbols & Abbreviations

Symbol/Index	Meaning/Abbreviation
NRM_s	Negative refractive index metamaterial
ENZ	Epsilon near zero
MNZ	mu-near-zero
EMNZ	Epsilon-and-mu-near-zero
ZIM	Zero index metamaterial
TCO	Transparent conducting oxide
SiC	Silicon Carbide
NZI	Near Zero Index
THG	Third Harmonic Generation
FWM	Four Wave Mixing
UV	Ultraviolet
EMT	Effective Medium Theory

CHAPTER 1
The Origin of Refractive Index

We've already stated that light travels slower in water than in air, and somewhat slower in air than in vacuum. The index of refraction n describes this phenomenon. Now we'd like to know how such a slower velocity might have occurred. We should look at the relationship between several physical assumptions or claims, such as

1. That the entire electric field in each physical situation may always be expressed by the total of all the charges in the universe's fields

2. A single charge's field is always given by its acceleration multiplied by a retardation at speed c.

While it is true that light or any electrical wave appears to travel at the speed c/n through a material with an index of refraction of n, the fields are still produced by the motions of all the charges—including the charges moving in the material—and with these fundamental contributions of the field travelling at the ultimate velocity c, we can see how the apparent slower velocity is achieved.

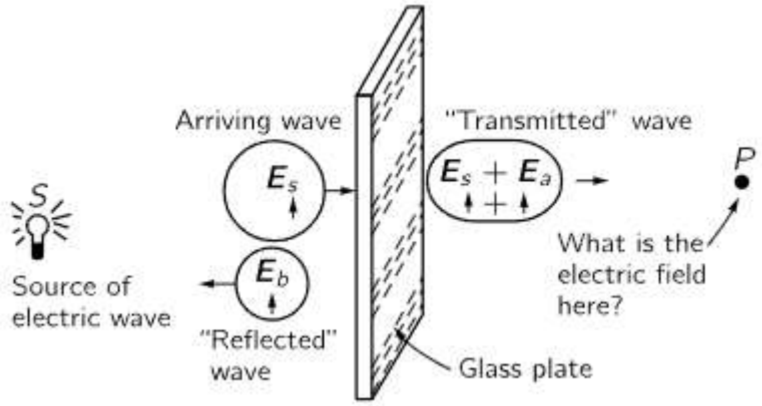


Fig 1. Electric waves passing through a layer of transparent material.

S and P are thought to be far distant from the plate in this example. An electric field anywhere that is far from all moving charges is the (vector) sum of the fields produced by the external source (at S) and the fields produced by each of the charges in the plate of glass, each with its proper retardation at the velocity c, according to the principles we stated earlier.

$$E = \sum_{all\ charges} E_{each\ charge} \tag{1}$$

$$E = E_s + \sum_{all\ \ other\ charges} E_{each\ charge} \tag{2}$$

where Es is the field owing only to the source and would be exactly the field at P if no material were present. If there are any additional moving charges, we anticipate the field at P to be different from that at Es. Consider the case where the impacts of the other atoms are negligible in comparison to the effects of the source. To put it another way, we take a material where the total field is not greatly affected by the mobility of the other charges. This relates to a material with a refraction index extremely near to one. The field produced at P by all the oscillating charges in the glass plate must be calculated.

This section of the field will be referred to as Ea, and it is just the second word expressed as a sum. We'll have the whole field at P when we add it to the term Es because of the source. If the total field at P is going to appear like radiation from the source that is slowed while travelling through the thin plate, the "correction field" Ea would have to be. A wave going to the right (along the z-axis) would have no impact if the plate had no influence on it.

$$E_s = E_0 \cos w(t - \tfrac{z}{c}) \tag{3}$$

$$E_s = E_0 e^{-i\omega(t-\frac{z}{c})} \tag{4}$$

The thickness of the plate Δz. If the plate were not there the wave would travel the distance Δz in the time Δz/c. But if it appears to travel at the speed c/n then it should take the longer time nΔz/c or the additional time Δt = (n−1)Δz/c. It would then resume travelling at the speed of c. We can account for the extra time it takes to pass through the plate by substituting t with (t−Δt) or by [t−(n−1)Δz/c]. As a result, the wave should be written after the plate is inserted.

$$E_{afterplate} = E_0 e^{i\omega\left(t-(n-1)\frac{\Delta z}{c}-z/c\right)} \tag{5}$$

$$E_{afterplate} = E_0 e^{-i\omega\left((n-1)\frac{\Delta z}{c}\right)} E_0 e^{i\omega\left(t-\frac{z}{c}\right)} \tag{6}$$

When Δz is tiny, it is quite simple to identify the proper amount to add; we will remember that if x is a small number, then e^x is nearly equal to (1+x). We can write, therefore

$$e^{-i\omega\left((n-1)\frac{\Delta z}{c}\right)} \approx 1 - i\omega\left((n-1)\frac{\Delta z}{c}\right) \tag{7}$$

$$E_{afterplate} = \underbrace{E_0 e^{i\omega}\left(t - \frac{z}{c}\right)}_{E_s} - \underbrace{\frac{i\omega(n-1)\Delta z}{c} E_0 e^{i\omega}\left(t - \frac{z}{c}\right)}_{E_a} \tag{8}$$

The field Es will have the same phase everywhere on the plate if the source S is far to the left, thus we may write that in the plate's vicinity.

$$E_s = E_0 e^{i\omega(t - z/c)} \quad (9)$$

$$E_s = E_0 e^{i\omega t} \ \ (at\ the\ plate) \quad (10)$$

When it comes to light difficulties, the right description of an atom provided by wave mechanics states that the electrons behave as if they were held by springs. Assume that the electrons have a linear restoring force that, when combined with their mass m, causes them to act like little oscillators with a resonant frequency of 0. We've already looked at such oscillators and know that their motion is described by the following equation:

$$F = m\left(\frac{d^2x}{dt^2} + \omega_0^2\, x\right) \quad (11)$$

F stands for the driving force.

The driving force in our problem comes from the electric field of the wave from the source, thus we should employ that.

$$F = q_e E_s = q_e E_0 e^{i\omega t} \quad (12)$$

The electron's equation of motion is then

$$q_e E_0 e^{i\omega t} \ = m\left(\frac{d^2x}{dt^2} + \omega_0^2\, x\right) \quad (13)$$

$$x = x_0 e^{i\omega t} \quad (14)$$

$$x_0 = \frac{q_e E_0}{m(\omega_0^2 - \omega^2)} \quad (15)$$

so that

$$x_0 = \frac{q_e E_0}{m(\omega_0^2 - \omega^2)} e^{i\omega t} \quad (16)$$

We know that

$$\text{total field at P} \ = \ -\frac{\eta q}{2\epsilon_0 c} i\omega x_0 e^{i\omega(t - \frac{z}{c})} \quad (17)$$

or just putting x_0 in above equations we get

$$E_a = \frac{\eta q}{2\varepsilon_0 c}\left[i\omega \frac{q_e E_0}{m(\omega_0^2 - \omega^2)} e^{i\omega(t-\frac{z}{c})}\right] \tag{18}$$

The two expressions of E_a will, in fact, be identical if

$$(n-1)\Delta z = \frac{\eta q_c^2}{2\epsilon_0 m(\omega_0^2 - \omega^2)} \tag{19}$$

Substituting NΔz for η and cancelling the Δz, we obtain our major result: a formula for the index of refraction in terms of the characteristics of the material's atoms—as well as the frequency of light:

$$n = 1 + \frac{N q_c^2}{2\epsilon_0 m(\omega_0^2 - \omega^2)} \tag{20}$$

This equation gives the "explanation" of the index of refraction that we wished to

obtain. In this formula we don't know about the natural frequency of atoms. And definitely we

can't get a general formula for the n. Because the value of ω0 is different for every different material. Now we discuss our formula in various possible circumstances.

2.1 For gases

The electron oscillators' inherent frequencies correlate to ultraviolet light, which means that 0 is substantially greater than visible light. The index for gases is basically constant. However, if we look attentively, we can observe that n climbs slowly with.

That is why a prism bends light in the blue direction more than in the red.

• If the frequency is very near to natural frequency or if natural frequency is evident. Because the denominator is zero, the index might grow exponentially.

• If is larger than the natural frequency, our equation is now negative, implying that n has a negative value. This indicates that the effective speed of the waves in the material exceeds c.

The speed at which you may transmit a signal, on the other hand, is defined not by the index at one frequency, but by the index at several frequencies. The index indicates the speed at which the wave's nodes travel. A wave's node is not a signal in and of itself. To convey a signal, you must alter the wave in some way, for as by cutting a notch in it or making it slightly thicker or thinner. That means the wave must have several frequencies, and it can be demonstrated that the speed at which signals move is determined not by the index alone, but by the way the index varies with frequency.

We need to make some adjustments to be entirely correct.

1. We should anticipate that our atomic oscillator model will have some dampening force.

2. A second change was made to account for the fact that each type of atom has many resonance frequency.

It's simple to solve our dispersion problem by thinking that there are multiple distinct types of oscillators, each acting independently.

CHAPTER 2.
Drude Model: Deriving the permittivity of metal

2.1 Derivation of electron motion

Let's say the driving oscillating electric field is E=E0 Cos(-ωt) The damping force, which is defined by the damping coefficient γ, is velocity dependant. (unit of chosen such that force = γmv)

$$\frac{d^2x}{dt^2} + \gamma\frac{dx}{dt} + \omega_0^2 x = \frac{qE_0}{m}\cos(-\omega t) \tag{21}$$

Solution of the equation $x = x_0\cos(-\omega t + \phi)$ (22)

In complex form, $x = x_0 e^{i\phi} e^{-i\omega t}$ (23)

$$\tilde{x} = \widetilde{x_0} e^{-i\omega t} \tag{24}$$

Take derivative

$$\frac{d\tilde{x}}{dt} = (-i\omega)\widetilde{x_0} e^{-i\omega t} \tag{25}$$

$$\frac{d^2\tilde{x}}{dt^2} = (-i\omega)^2\widetilde{x_0} e^{-i\omega t} \tag{26}$$

Put all of these numbers into equation 1 and solve for the complex form amplitude of electron motion.

$$\widetilde{x_0} = \frac{qE_0}{m}\frac{1}{(\omega_0^2 - \omega^2 - i\omega\gamma)} \tag{27}$$

The fact that it's complicated simply implies that there's a time delay (phase shift) between the generating electric field and the electron's reaction.

$$\widetilde{x_0} = \frac{qE_0}{m}\frac{1}{(\omega_0^2 - \omega^2 - i\omega\gamma)} e^{-i\omega t} \tag{28}$$

We obtain after finding the real and imagined parts.

$$x_{actual}(t) = \frac{qE_0}{m}\frac{\omega_0^2 - \omega^2}{(\omega_0^2 - \omega^2)^2 + (\omega\gamma)^2)}\cos(-\omega t + \phi) \tag{29}$$

2.2 Dipole moment

The complex dipole moment created by an electron travelling like this in an atom (with the nucleus at the origin, immobile and hence not contributing to the dipole moment) is calculated as follows:

$$\boldsymbol{P} = \sum \boldsymbol{q_i}\, \boldsymbol{r_i} \tag{30}$$

$$\tilde{x} = (q)\left(\frac{qE_0}{m}\frac{1}{(\omega_0^2-\omega^2-i\omega\gamma)}e^{-i\omega t}\right)x \tag{31}$$

$$\tilde{x} = \frac{q^2E_0}{m}\frac{1}{(\omega_0^2-\omega^2-i\omega\gamma)}e^{-i\omega t} \tag{32}$$

The polarisation P stands for the dipole moment per volume. If there are γN electrons per volume, each operating with the same dipole moment as described above, the complex polarisation is obtained by multiplying the last equation by N.

$$\tilde{P} = \frac{Nq^2E_0}{m}\frac{1}{(\omega_0^2-\omega^2-i\omega\gamma)}e^{-i\omega t} \tag{33}$$

$$\tilde{P} = \frac{Nq^2}{m}\frac{1}{(\omega_0^2-\omega^2-i\omega\gamma)}(E_0e^{-i\omega t}) \tag{34}$$

$$\tilde{P} = \frac{Nq^2}{m}\frac{1}{(\omega_0^2-\omega^2-i\omega\gamma)}\tilde{E}$$

Between P and E, there is a phase change.

2.3 Susceptibility and Permittivity

Because susceptibility is determined by P=$\epsilon_0\chi E$ We can just read off the susceptibility from the previous equation as the material multiplying Ē divided by

$$\tilde{x} = \frac{Nq^2}{m\epsilon_0}\frac{1}{(\omega_0^2-\omega^2-i\omega\gamma)} \tag{35}$$

Plasma Frequency is a constant number that also happens to be the frequency at which a plasma will spontaneously oscillate if the positive and negative charges in the plasma are offset from each other.

$$\omega_P = \sqrt{\frac{Nq^2}{m\epsilon_0}} \tag{36}$$

As a result, we may write susceptibility in a very concise way,

$$\tilde{x} = \frac{\omega_P^2}{(\omega_0^2-\omega^2-i\omega\gamma)} \tag{37}$$

Furthermore, because susceptibility and permittivity (dielectric constant) are linked,

$$\epsilon_r = 1 + \chi \tag{38}$$

$$\bar{\epsilon_r} = 1 + \frac{\omega_P^2}{(\omega_0^2-\omega^2-i\omega\gamma)} \tag{39}$$

At a very low frequency value ω=0

$$\bar{\epsilon_r}(DC) = 1 + \frac{\omega_P^2}{(\omega_0^2 - 0^2 - i0\gamma)} \quad (40)$$

$$\bar{\epsilon_r}(DC) = 1 + \frac{\omega_P^2}{(\omega_0^2)} \quad (41)$$

And high frequency value

$$\bar{\epsilon_r}(high\ frequency) = 1 + \frac{\omega_P^2}{(\omega_0^2 - \infty^2 - i\infty\gamma)} \quad (42)$$

$$\bar{\epsilon_r}(high\ frequency) = 1 \quad (43)$$

CHAPTER 3
Zero Refractive Index

3.1 Negative Refractive Index

Despite the fact that it was known that the refractive index would have to be a complex number to account for absorption and even a tensor to characterise anisotropic materials, the sign of the refractive index was never addressed. Veselago originally addressed the scenario of a medium with both negative dielectric permittivity and negative magnetic permeability at a particular frequency in 1967, and came to the conclusion that the medium has a negative refractive index. Despite the fact that Veselago went on to point out several interesting effects in NRMs, such as a modified Snell's law of refraction, a reversed Doppler shift, and an obtuse angle for Cerenkov radiation, his result remained an academic curiosity for a long time because real materials with simultaneously negative and were not available. However, theoretical concepts for structured photonic media with negative and in particular frequency ranges have been established experimentally in recent years, bringing Veselago's result to the forefront.

The effective refractive index is negative. The complex refractive index of a medium is the ratio of the speed of an electromagnetic wave through that medium to the speed of an electromagnetic wave in vacuum, and it may be used to calculate relative dielectric permittivity. If both ε and μ are negative in a given thus be written as n =$\sqrt{|\mu||\varepsilon|}$, where μ is complex relative magnetic permeability and ε wavelength range, this means that we may write μ = |μ| exp(iπ) and in an equivalent fashion ε = |ε| exp(iπ). It follows that

$$n = \sqrt{|\mu||\varepsilon| \exp(2i\pi)} \qquad (44)$$

$$n = -\sqrt{|\mu||\varepsilon|} \qquad (45)$$

NRM metamaterial is a combination of two materials that independently demonstrate negative permeability and permittivity, as no known material has these properties intrinsically. ε<0 and μ<0.

The lossy Drude model, in which polarisation and magnetization are separated, is a commonly used form for effective and.

$$\varepsilon_{eff}(\omega) = 1 - \frac{\omega_{pe}^2}{\omega(\omega+i\Gamma_e)} \qquad (46)$$

$$\mu_{eff}(\omega) = 1 - \frac{\omega_{pm}^2}{\omega(\omega+i\Gamma_m)} \qquad (47)$$

For both polarisation and magnetization, the plasma frequency ωp and the damping constant Γ are commonly considered to be identical,

$$\omega_{pe} = \omega_{pm} = \omega_p, \quad \Gamma_{pm} = \Gamma_{pe} = \Gamma_p \tag{48}$$

The refractive index of a low loss model becomes

$$n_{eff}(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega+i\Gamma)} \approx 1 - \frac{\omega_p^2}{\omega^2} + i\Gamma\frac{\omega_p^2}{\omega^3} \tag{49}$$

The assumption that dumping is negligible is a common one, and it's a good estimate even from an experimental standpoint. Finally, if lossy metamaterial is explicitly considered, the damping factor is usually expressed as a proportion of plasma frequency. The Lorentz model, which is relevant to various experimental implementations of negative permeability structures, is another model frequently seen in literature.

3.2 Zero Refractive Index

Near zero material parameters are not a new phenomena, as we all know. Metamaterial is classed as epsilon-near-zero (ENZ), mu-near-zero (MNZ), or epsilon-and-mu-near-zero (EMNZ) depending on whether permittivity, permeability, or both are zero. All of these metamaterials have a zero index (ZIM). EMNZ metamaterials, on the other hand, have been renamed ZIM metamaterials[2]. ZIM is the only one with both $\varepsilon = 0$ and $\mu = 0$. The zero refractive index allows for a wide range of applications. Waveguide Rectangular This feature has always been present in rectangular waveguides, long before metamaterial was recognised. Consider a rectangular waveguide. The wave number k for TEmn mode is given by, where m index refers to the bigger dimension a and n index relates to the smaller dimension b.

$$\kappa = \sqrt{\left(\frac{\omega}{c}\right)^2 - \pi^2\left[\left(\frac{m}{a}\right)^2 + \left(\frac{n}{b}\right)^2\right]} \tag{50}$$

$$\kappa = \frac{1}{c}\sqrt{\omega^2 - \omega_{mn}^2} \tag{51}$$

$$\omega_{mn}^2 = \left[\left(\frac{m}{a}\right)^2 + \left(\frac{n}{b}\right)^2\right] \tag{52}$$

is the frequency of the cut-off. The wave velocity, also known as phase velocity, is expressed as

$$v_p = \frac{\omega}{k} = \frac{c}{\sqrt{1-\omega_{mn}^2/\omega^2}} \tag{53}$$

$$v_g = \frac{d\omega}{dk} = c\sqrt{1 - \omega_{mn}^2/\omega^2} \tag{54}$$

and the group velocity is given by Below cut-off frequency ($\omega < \omega_{mn}$), no mode exists as k is imaginary and for very high frequencies ($\omega >> \omega_{mn}$), the propagation resembles that in free space. At extremely high frequencies, the wavelength λmn is so tiny in comparison to the dimensions a and b that the incoming wave seems to be moving in empty space. The TE_{10} mode corresponds to the lowest cutoff frequency of ω_{10}. At $\omega = \omega_{mn}$, the wave vector k is 0 for any mode, hence the effective refractive index is zero and the phase velocity is infinite, as it should be It should be emphasised that if phase velocity exceeds c, or even infinite, no physical rules are broken because it has no physical relevance. The group velocity is the rate of energy transmission that is always smaller than c. As a result, it has been established that a physical system's zero refractive index is a highly natural attribute.

3.3 Snell's Law for NZI Material

(1) If n1 is zero :-

If light travels from zero refractive index to any medium with refractive index n_2 then

$n_1 = 0$

$n_2 \sin(r) = 0, n_2 \neq 0$

So only,

$\sin(r) = 0$

The refraction angle (r) is thus always zero.

The angle of refraction in the second media is always zero or the transmitted light is directed towards the normal for any angle of incidence in the first medium.

(2) If n2 is zero:

If light travels from a media having a refractive index of n1 to a medium with a refractive index of zero,

$$n_1 \sin i = n_2 \sin r \qquad , \; n_2 = 0$$

$$n_1 \sin i = 0$$
$$\sin i = 0$$

As a result, the light must incident normally before passing through ZIM.

CHAPTER 4.
ENZ Material

4.1 Classifications

"Materials with a permittivity (ENZ) that approaches zero" The ENZ regime occurs in general in a wavelength range where both the real and imaginary parts of the permittivity are near to zero. Epsilon-near-zero (ENZ) materials are intriguing nonlinear plasmonic materials because of their unique optical characteristics. Several transparent conductive oxides (TCOs) have been found to have substantial optical nonlinearities at their ENZ wavelengths, including indium-tin oxide (ITO) and aluminum-doped zinc oxide (AZO)[4]. Furthermore, ENZ modes of ENZ materials thin films may be used to couple with plasmonic resonant modes to increase optical modulation efficiency. ENZ materials have long been thought of as a key platform for integrated devices and nanophotonics.

In a number of applications, such as nonlinear optical switching and mode coupling, a material's ENZ wavelength must adjust to a desired working wavelength. "Materials having ENZ in the wavelength range of 1525nm – 1565nm are particularly appealing for optical telecommunications applications."

Because of its potential for low-cost production and compatibility with silicon-based integrated photonics, ITO has shown special promise among these materials. In commercial semiconductor integrated circuits, ITO is frequently employed. As a result of its compatibility with well-established integrated photonics production procedures, it is an appealing material for building ENZ-based optoelectronic devices. Furthermore, it has been shown that annealed ITO has a quicker recombination time than many semiconductors.

4.2 Indium Tin Oxide (ITO)

Because of its unique optical characteristics, indium tin oxide (ITO) offers a wide range of uses at its epsilon-near-zero (ENZ) wavelength. The ENZ wavelength can be easily tuned by post-annealing. We show that thermal annealing in air for up to 130 minutes at 330°C may red-shift the ENZ wavelength of ITO films over a wide range of wavelengths from 1200 nm to 1550 nm. The Drude model parameters of plasma frequency, damping factor, electron mobility, and effective mass were extracted using optical transmittance and reflectance spectra, as well as electron densities, for these ITO samples. The findings reveal that the red-shift in plasma frequency and ENZ wavelength is caused by changes in electron density and effective mass. At its epsilon-near-zero (ENZ) wavelength, indium tin oxide (ITO) has a wide range of applications due to its unique optical properties. Post-annealing allows for easy tuning of the ENZ wavelength. Thermal annealing in air for up to 130 minutes at 330°C red-shifts the ENZ wavelength of ITO films throughout a wide range of wavelengths from 1200 nm to 1550 nm, according to our findings. Optical transmittance and reflectance spectra, as well as electron densities, were used to derive the Drude model parameters of plasma frequency, damping factor, electron mobility, and effective mass for these ITO samples. Changes in electron density and effective mass produce the red-shift in plasma frequency and ENZ wavelength, according to the findings.

- Indium tin oxide (ITO) is a ternary compound with variable amounts of indium, tin, and oxygen. Indium tin oxide is commonly found in an oxygen-saturated state, having a formula of 74 percent indium, 18 percent tin, and 8% oxygen by weight.
- It is translucent and colourless in thin layers, but yellowish to orange in bulk grey.
- It serves as a metal-like mirror in the infrared portion of the spectrum.
- Indium tin oxide is one of the most widely used transparent conducting oxides because of its electrical conductivity and optical transparency, as well as the ease with which it can be deposited as a thin film.
- As with all transparent conducting films, a compromise must be made between conductivity and transparency, since increasing the thickness and increasing the concentration of charge carriers increases the film's conductivity, but decreases its transparency

CHAPTER - 5
ENZ Properties and Applications

The interplay of light and matter has sparked several scientific breakthroughs during the last decade. Recently, the spectral area where the material's index of refraction approaches 0 has attracted curiosity, resulting in a variety of exciting phenomena such as static light, enhanced non-linearities, and so on. Near-zero material parameters are not a relatively new development; metals have 0 permittivity at plasma frequency, while polaritonic materials such as silicon carbide (SiC) have zero permeability [3]. However, we can obtain near-zero optical characteristics in a more controlled and varied manner with metamaterial.
Structured materials can only obtain NZI behaviour as an effective property that happens only at distances bigger than the size of structural units, whereas naturally occurring NZI materials accomplish this property locally. This may also be done in thin films with low losses that can be combined with other structures like metasurfaces and guiding structures. However, when the impedance approaches 0 or infinity, a significant restriction of homogeneous materials is the impedance mismatch that occurs when producing low-loss NZI materials.
The NZI condition has been demonstrated in a variety of systems, including homogeneous materials, metals, doped semiconductors, and photonics materials (structured materials such as metamaterials), waveguides with near-zero cut-off frequencies, resonant cavities, and photonic crystals.

Depending on whether the permittivity, permeability, or both are zero, the material is characterised in several ways. Category names include epsilon-near-zero (ENZ), mu-near-zero (MNZ), and epsilon-and-mu-near-zero (EMNZ) [2]. All of these metamaterials are technically zero index metamaterials. According to the literature, the ZIM is the one that equals both 0 and 0. It paves the way for a slew of new optical phenomena, including photon tunnelling, super coupling [6-8], emission control, and severe nonlinear interactions. All of these materials have several characteristics that make NZI effects unique. as discussed below, including divergent velocities, wavelength expansion, and electric field enhancement .

ENZ Properties and application

1. Diverging velocity- We investigate the propagation of a plane wave in an area defined by a Drude oscillator to demonstrate the phenomenon of diverging velocities in a material with a refractive index near to zero. As $\omega \rightarrow \omega_p$ in the lossless case, where ω_p is the plasma frequency, $n(\omega) \rightarrow 0$ .Thus the phase velocity diverges $v_p(\omega)= c/\sqrt{\varepsilon(\omega)}$ and $\varepsilon = n^2/\mu$[12]

$$v_g(\omega) = d\omega/dk = \frac{c\sqrt{\varepsilon(\omega)}}{\varepsilon(\omega)+(\omega/2)(d\varepsilon(\omega)/d\omega)} \qquad (55)$$

Because the slope of permittivity is limited[9-12], it tends to zero.

Diverging phase velocities are a key property of NZI materials since they boost light-matter interaction significantly. They've also been investigated in a variety of optical applications, including sensors, cloaking, and transformation optical devices, near-perfect optical absorbers, optical storage systems with enhanced quantum and nonlinear interactions, and pulse shaping.

2. Wavelength expansion

The obvious consequence of diverging phase velocity in NZI material is wavelength expansion.

$\lambda_{NZI} = \lambda_0/n$ where λ_{NZI} Is the wavelength at which n approaches zero λ_0 is the free space wavelength**.**

As a result, when the refractive index n is low, the wavelength grows dramatically, and phase advance is essentially non-existent. ($kL = 2\pi L/\ \lambda_{NZI} \rightarrow 0$) Wave propagating through a NZI film with a thickness L substantially greater than the wavelength in open space This diverging wavelength effect has been exploited to make phase-matching-free nonlinear optical devices[13-15], as well as super-coupling[16-18], antenna resonance pinning[19-21], and geometric resonance cavities.

It was also the subject of some fascinating theoretical work on 'DC' or'static' light.

3. Field enhancement

We know that an electromagnetic boundary condition causes electric field increase in a material. This necessitates the application of the rule to the normal component electric field across a boundary $\varepsilon_1 E_1^{\perp} = \varepsilon_2 E_2^{\perp}$. This resulted in an increase in the E's normal component within the NZI material, $E_2^{\perp}$, by a factor of $\varepsilon_1/\ \varepsilon_2$. Because $\varepsilon_2 \rightarrow 0$, It is possible to achieve effective energy confinement within the NZI layer.

APPLICATION

The NZI material has been used in a variety of applications, including electromagnetic waveguides, free space wave manipulations, metrology, non-linear optics, lasers, and quantum optics.

We'll be concentrating on nonlinear optics here. Using natural material non-linearities, photon-photon interaction can be improved. In this discipline, a lot of work is put into developing new techniques to increase nonlinear effects and inventing or manufacturing materials that display nonlinear reactions.

Because of their modest refractive index and the features outlined above, NZI materials are highly suited for improving non-linear optical interaction.

Many applications in modern photonics, such as telecommunications and all optical data processing and storage, spectroscopy, and quantum information technologies, need non-linear optical interaction. The efficiency of nonlinear optical interaction is determined by several essential factors, including phase matching, photon interaction length, and peak electric field. NZI materials can assist achieve these requirements.

Now we'll look at some non-linear qualities.

CHAPTER - 6
Non-Linear properties and application

Non-linear Properties and applications

1. *Third order susceptibility*

ENZ materials' strong nonlinear optical response is linked to increases in nonlinear susceptibility and the electric field that occurs within the low permittivity material.

From the preceding equation, we may derive higher order susceptibilities $\chi^{(3)}$, which are a function of wavelength.

$$n(I) = \sqrt{\varepsilon}(E) = \sqrt{\varepsilon^{(1)} + 3\,|E\,|^{(2)}\chi^{(3)}} \qquad (56)$$

The value of $\chi^{(3)}$ is found to be higher in a limited area surrounding the zero-permittivity wavelength. When comparing qualities at zero permittivity wavelength to those at other wavelengths. The value of third order susceptibility is claimed to stay constant, which cannot explain for the size of the rise in non-linear optical response. So, for a given incident pump field intensity, we predict a field augmentation mechanism inherent to ENZ material to greatly boost |E|.

2. *Continuity of electric field*

The ENZ material's low permittivity results in a unique field augmentation process. In the absence of surface charge, the normal component of the electric displacement field remains constant.Thus, we get $|E_\perp = \varepsilon^{-1}|E_{0,\perp}|$. We get the total field within the medium of permittivity when a p polarised beam is incident at an angle.

$$E = |E_0|\sqrt{\cos^2\theta + \frac{\sin^2\theta}{\varepsilon}} \qquad (57)$$

When seen from an oblique angle, the electric field within an ENZ medium might be much larger than the incident field. As a result, many non-linear effects are angularly dependent.

3. *Slow light nonlinear enhancement*

For ENZ medium the group velocity is $v_{g=}\sqrt{\varepsilon}c$. Thus, it will feature an asymptotically vanishing group velocity as $\varepsilon \rightarrow 0$. The ENZ mode does, in fact, exhibit a diminishing group velocity. Because slow light propagation has been linked to nonlinear amplification in the past, researchers have drawn a link between ENZ-based nonlinearities and slow light effects.

4. *ENZ modes and Barreman modes*

An ENZ thin film has a unique set of propagating eigenmodes, such as unbounded Brewster or Berreman mode and the restricted ENZ mode. The latter has a flat dispersion profile and a significant field augmentation thanks to the boundary condition. A thin coating must be ultra-thin to support ENZ.

NON-LINEAR APPLICATION

Harmonic generation and frequency mixing are two of the most common uses of nonlinear optical phenomena. Multiple theoretical works had predicted an increase in harmonic generation in ENZ and NZI media before experiments, but it has now been demonstrated that in a strongly resonant system loaded with ITO and operating under ENZ conditions, both the second-order nonlinear response from free electron and the third-order nonlinear optical response due to bound electron contribute to third harmonic generation (THG).
Through the FWM (Four Wave Mixing) procedure, a set of possible frequency-mixing possibilities is also accessible based on the idea of third order non-linearities. The efficiency is particularly sensitive to the phase-matching criteria and the refractive index of the material since three input photons are involved. Because of the NZI media's divergent wavelengths, phase matching constraints are substantially loosened, and all photons add coherently.

CHAPTER 7

RESULT AND DISCUSSION

We looked into the theoretical description of a composite nonlinear optical material made up of two layers with varying linear and nonlinear properties. Unit layers of Ag and SiO_2 were used to produce the multilayer ENZ structure. The Ag thickness in each layer is kept at 5 nm in order to achieve ENZ conditions around 820-890 nm in the centre of the laser tuning zone, while the SiO_2 thickness is 70 nm. The optical properties of a composite material may differ significantly from those of its constituents. We use the programming and numeric computation platform MATLAB to theoretically compute all of the linear and non-linear parameters in our proposed study.

Figure 1. The multilayer ENZ structure was made up of unit layers of Ag and SiO_2. The thickness of the Ag and SiO2 layers is represented by t_m and t_d, respectively.

2.1. Linear Response

The dielectric constant ε_m may be represented as a function of frequency ω using the Drude model.

$$\varepsilon_m(\omega) = \varepsilon_{m,r} + i\varepsilon_{m,i} = \varepsilon_b - \frac{\omega_p^2}{\omega^2+\gamma^2} + \frac{i\gamma\omega_p^2}{\omega^3+\gamma^2\omega} \quad (58)$$

Where $\varepsilon_{m,r}$ and $\varepsilon_{m,i}$, are the real and imaginary components of the dielectric constant of the metal, respectively. ε_b is the static term owing to bound charge and γ is the damping coefficient.

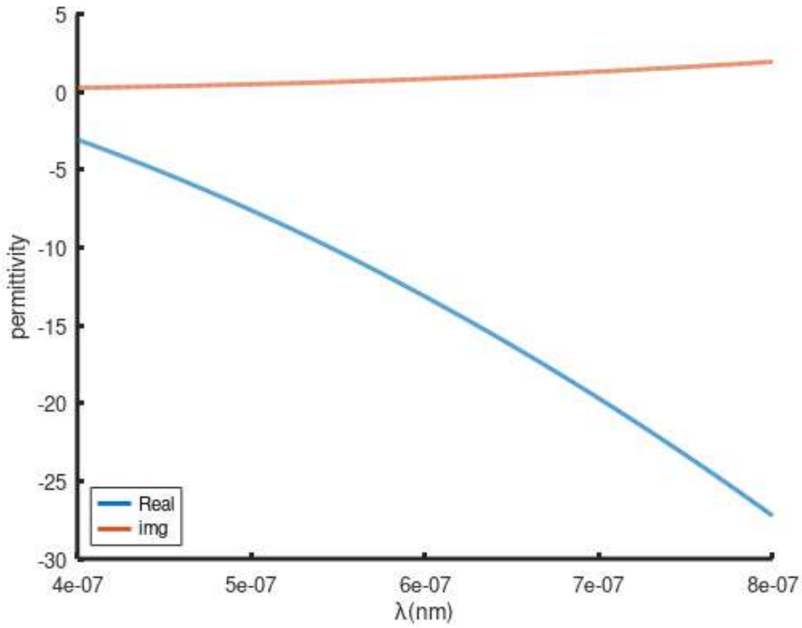


Figure 2. Variation of permittivity with wavelength for Ag

Negative dielectric constant metals, such as Ag, pass through zero before becoming positive at plasma frequencies, which are frequently UV. To get a near-zero dielectric constant at lower frequencies, such as visible light, we must basically "dilute" the metal with a positive dielectric constant material like SiO2 . Negative permittivity Ag loses less in the visible region than most noble metals, but positive permittivity SiO2 has good thermal stability and negligible optical loss. Unit layers in the proposed multilayer ENZ structure have sub-wavelength thicknesses to fulfil the EMT criterion. In addition, the thickness of the Ag layer must be kept as low as possible to reduce the overall structure's optical loss.

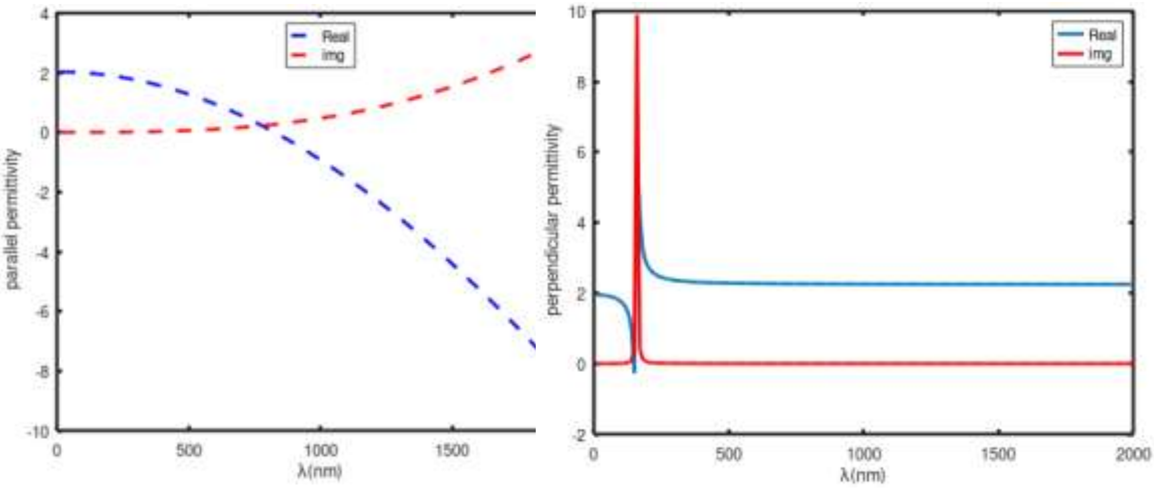
Maxwell Garnett was the first to analyse composite materials theoretically, considering the linear response of metallic inclusion particles contained in glass and explaining the colours of metallic colloid. The effective permittivity of the multilayer may be calculated using the Maxwell-Garnett formulae. The permittivity components parallel to the luminant layer can be expressed as

$$\varepsilon_{\parallel} = \rho\varepsilon_m + (1-\rho)\varepsilon_d \qquad (59)$$

When the electric field is perpendicular to the layer plane, The electric field, on the other hand, is not evenly distributed over the two composite components, and the effective linear dielectric constant is given by

$$\frac{1}{\varepsilon_{\perp}} = \frac{\rho}{\varepsilon_m} + \frac{(1-\rho)}{\varepsilon_d} \qquad (60)$$

Where $\rho = t_m / (t_m + t_d)$.



(a) (b)

Figure 3. Permittivity of $AgSiO_2$ parallel and perpendicular component respectively. (a) The real part (dashed blue line) crosses zero at 826nm and imaginary Part (dashed red line) have small value for Ag(5nm) and SiO2(70nm) multilayer structure **(b)** solid blue line shows real part of perpendicular permittivity and solid red line shows imaginary part of perpendicular permittivity.

As a function of wavelength, Epsilon-Near-Zero metamaterials such as $AgSiO_2$ (thickness 70 nm) demonstrate a change in the real component of the dielectric permittivity from positive to negative values. For the given structure, ε′ is measured to be zero at 826nm (fig - 3a). The metamaterial's reaction may be expressed in terms of a conventional third order optical nonlinearity, which exhibits a clear inversion of the real and imaginary portions' roles upon crossing the ENZ wavelength, demonstrating an optically driven change in the metamaterial's physical behaviour.
A semiempirical equation based on the linear refractive index is used to derive the value of nonlinear refractive index/optical susceptibility. The linear optical susceptibility $\chi^{(1)}$ of a nonlinear medium that may be computed using the provided relation

$$\chi^{(1)} = \frac{(n^2 - 1)}{4\pi} \tag{61}$$

The thickness of the layers of material d and m might vary in general. In particular, if the electric field within the composite metamaterial is spatially uniform, the composite's optical constant becomes a simple average of its component materials, i.e,

$$\chi^{(1)} = \rho\chi_m + (1 - \rho)\chi_d \tag{62}$$

Because the electric field is not equally distributed between the two components, the composite's effective susceptibility might often exceed that of its constituent materials.

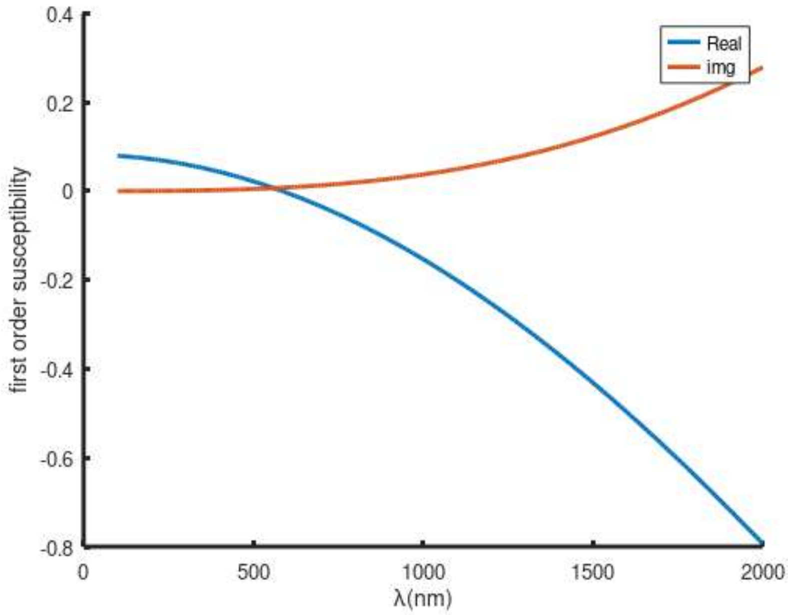


Figure 4. $\chi^{(1)}$ of $AgSiO_2$

. Linear susceptibility $\boldsymbol{\chi^{(1)}}$ has Similar variation as of linear permittivity. In terms of variation Light polarised parallel to the film interacts with an effective homogenised medium rather than each individual layer of the multilayer structure in the approximation of profoundly subwavelength films

1.2. Nonlinear Response

From miller's principles the third order susceptibility $\chi^{(3)}$ can be calculated

$$\chi^{(3)} = \frac{A(n^2-1)^4}{(4\pi)^4} \tag{63}$$

The constant A=1.7x10^{-10} esu, n is the refractive index which can be calculated by using the equation $\sqrt{\varepsilon}$ [9-10][15]. The composite is made up of layers of two distinct materials with linear dielectric constants that alternate. $\varepsilon_m$and ε_d and nonlinear susceptibilities $\chi_d^{(3)}$ and $\chi_m^{(3)}$,respectively

$$\chi^{(3)} = \rho\chi_d^{(3)} + (1-\rho)\chi_m^{(3)} \tag{64}$$

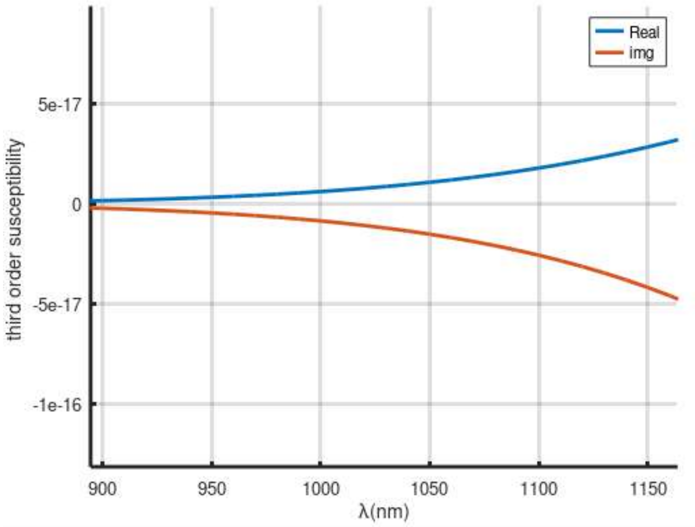


Figure 5 . Graph of $\chi^{(3)}$ show the real (solid blue line) and imaginary (solid red line) part of third order susceptibility, it exhibit a small increase near the value of ENZ

We can expect to see a region where ε_r is slightly negative in the vicinity of the ENZ wavelength if the nonlinear response is fast enough and the $\chi^{(3)}$ is positive (alternatively, for materials with negative $\chi^{(3),}$ identical results can be achieved at a wavelength with a slightly positive real permittivity).

The nonlinear refractive index can be calculated using the equation

$$n_{2r} = \frac{3[n_r \chi_r^{(3)} + n_i \chi_i^{(3)}]}{4\varepsilon_0 c n_r (n_r^2 + n_i^2)} \tag{65}$$

where A and B (representing $\chi_r^{(3)}$ and $\chi_i^{(3)}$, respectively) are used as criteria for fitting and A = 1.49×10^{-17} cm^2/V^2 and B = 1.9×10^{-17} cm^2/V^2.

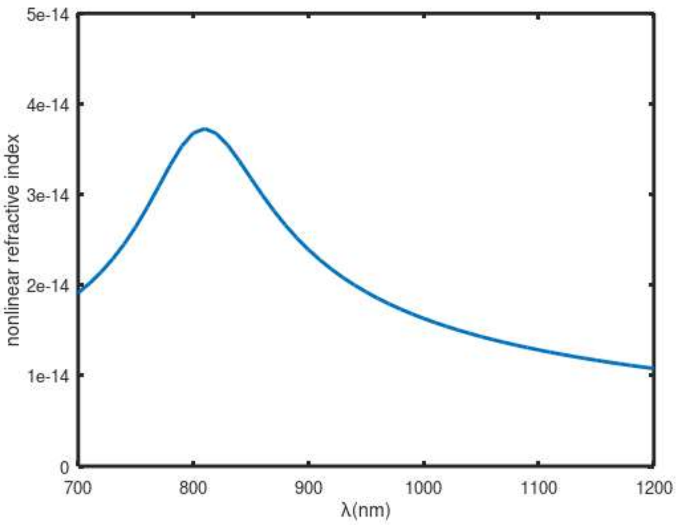


Figure 6. Graph of nonlinear refractive index

By requiring the longitudinal components of a TM-polarized field's displacement vector to be continuous across the border of media with different optical properties, the ENZ material improves the local electromagnetic field. For homogeneous, flat structures, this condition is expressed as $\varepsilon_{in}E_{in} = \varepsilon_{out}E_{out}$, where ε_{in} and ε_{out} are the dielectric constants inside and outside the medium, respectively, and E_{in} and E_{out} are the corresponding longitudinal components of the electric field amplitude, and the ENZ point is excited by oblique incidence. As a result, as ε_{in} declines, E_{in} rises, enhancing nonlinear optical phenomena such as the nonlinear index of refraction n_{2r}.

Most metals exhibit negative permittivity in the optical spectrum because their plasma frequencies are in the ultraviolet (UV). To "lower" the ω_p, it is preferred to use chemical reactions or doping methods to create metal-dielectric compounds. Positive permittivity dielectric components can push metal-dielectric compounds' effective permittivity into the ENZ and even positive areas. This phenomenon is a preferred technique to realise ENZ media since it lowers the ω_p of the compounds. This method, in particular, is used to build TCOs that demonstrate ENZ responsiveness.

The permittivity dispersion profile of metals (such as A_g) and degenerately doped semiconductors is based on the free electron represented by the Drude model.

$$\epsilon(\omega) = \epsilon_b - \frac{\omega_P^2}{\omega^2 + i\gamma\omega} \tag{66}$$

Where ϵ is the high-frequency permittivity, γ the electron damping term, and ω_P is the plasma frequency, as calculated by:

$$\omega_P = \sqrt{\frac{Ne^2}{m_e^* \epsilon_0}} \tag{67}$$

N is the free-electron volume density, and m_e^* is the electron's effective mass.

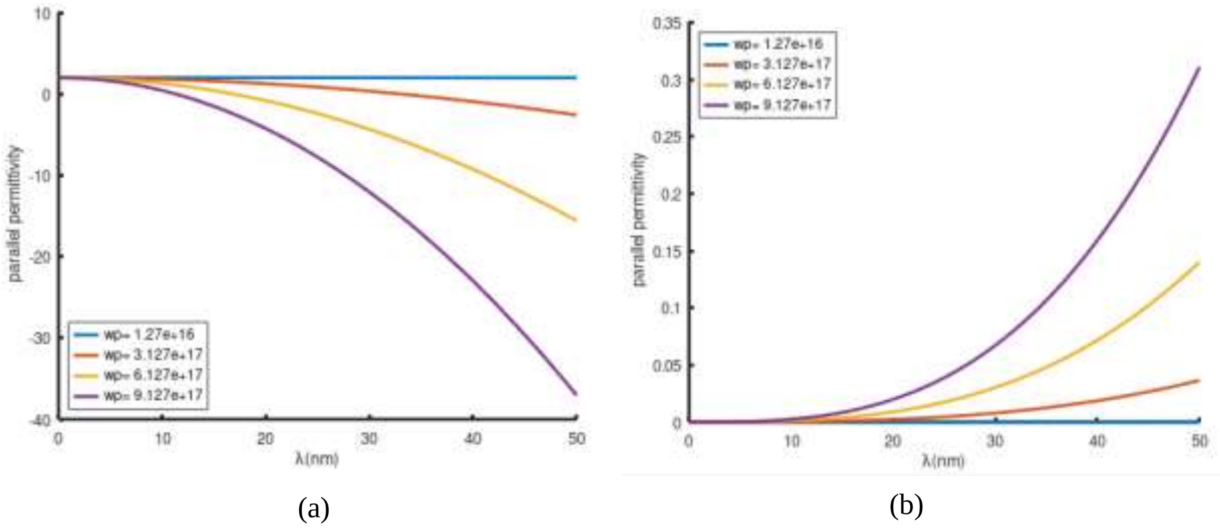


Figure 7. Variation in permittivity with wavelength at different ω_p (plasma frequency) (a) Real part (b) Imaginary part

An increase in carrier density causes a rise in plasma frequency, which reduces the real component of the permittivity, as represented by the preceding equation. In a doped semiconductor, the photon energy must exceed the bandgap energy; in a metal, it must exceed the interband transition energy.

Furthermore, by altering the thickness (t_d) of the SiO2 unit layer, we were able to modify the ENZ range of this Ag/SiO2 multilayer arrangement. At different wavelengths, the associated real component of the effective permittivity was zero for different thickness.

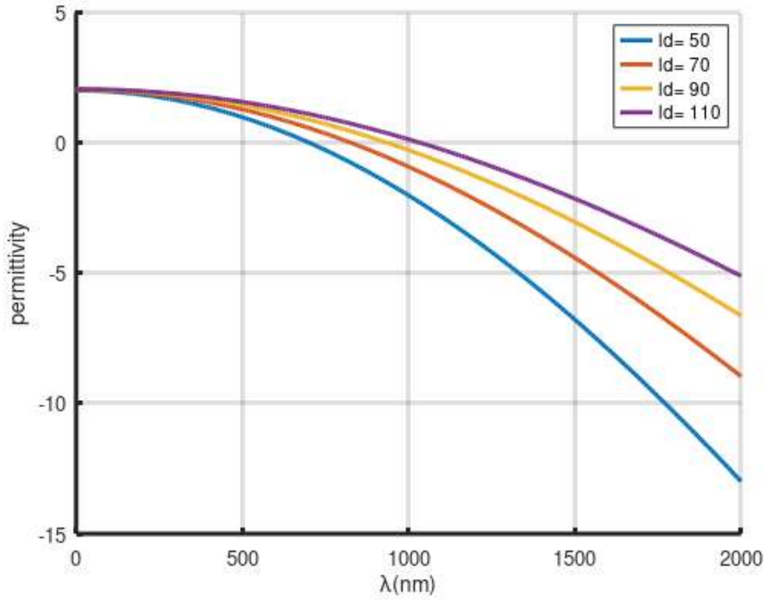


Figure 8. Variation of permittivity with l_d (dielectric thickness)

We have observed that if we increase the thickness l_d then real part of ε is increases and imaginary part will decrease along with the increase in λ_{ENZ}

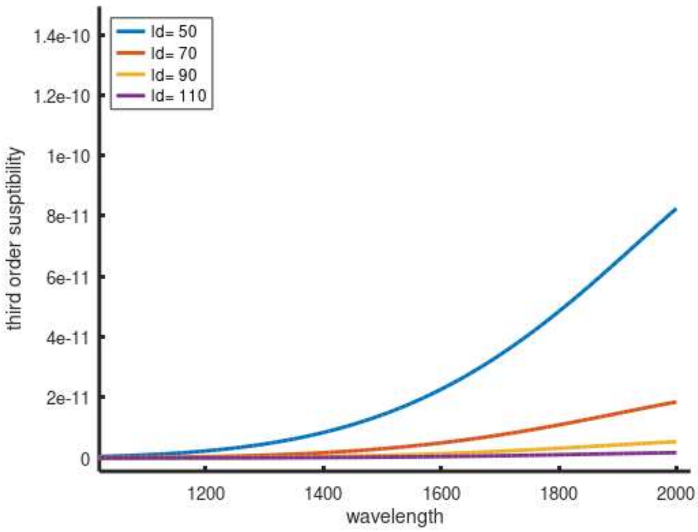


Figure 9. Variation of third order susceptibility with l_d (dielectric thickness) if we increase the thickness l_d the non-linear susceptibility $\chi^{(3)}$ will increase

CHAPTER 8

CONCLUSION

We have theoretically analysed the variation of permittivity (for metals such as Ag) with wavelength in our proposed study and concluded that to get a near zero dielectric constant, We must "dilute" the metal using a substance having a positive dielectric constant, such as SiO_2. In the near infrared range, the optical behaviour of a composite non-linear metamaterial ($AgSiO_2$) made up of alternating layers of fused silica glass and silver with thicknesses adjusted so that $\varepsilon'=0$ is theoretically explored. We also looked at the ($AgSiO_2$) transition from a positive to a negative value in the real component of the dielectric permittivity as a function of wavelength. At 826nm, ε' is measured to be zero for the given structure (thickness equal to 70nm). If the nonlinear response is fast enough and $\chi(3)$ is positive, we can expect to find a region where ε_r is relatively negative close to the ENZ wavelength. When a result, as in (the dielectric constant inside the medium) drops, E_{in} rises, and nonlinear optical phenomena, such as the nonlinear index of refraction n_{2r}, become more prominent.

The variation of ENZ wavelength with dielectric layer thickness and plasma frequency has also been investigated. We found that as the thickness of l_d is increased, the real part of ε is increases while the imaginary part decreases, corresponding to an increase in λ_{ENZ} and non-linear susceptibility $\chi^{(3)}$. We also noticed that as ω_p (plasma frequency) is increased, the ENZ value decreases, the real part of permittivity rapidly becomes negative, while the imaginary part remains positive.

this work can find application in photon generation via wave mixing process where high non-linearities are required which can be explored in future/ we expect our work will provide path for future developments in photon generation

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