

Nonlinear Real Index of Refraction Variations of a Gas Medium Due to a Monochromatic Radiation Near Resonance

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We have calculated the nonlinear real index of refraction variations of a gas medium due to a strong monochromatic radiation causing saturation effects. The gas is supposed to be composed of two-level molecules with which the external field is nearly resonant. We assume homogeneous (hard collisions, spontaneous decay) and inhomogeneous (Doppler effect) broadening mechanisms acting on the real index of refraction of the medium. The nonlinear dispersion of the medium is studied as a function of the detuning frequencies, saturation conditions and for various ratios between the homogeneous and inhomogeneous linewidths. In particular, the modification of the index of refraction due to saturation effects are emphasized.

Calculamos as variações do Índice de refração real não-linear de um meio gasoso devidas a uma radiação monocromática intensa que causa efeitos de saturação. Supõe-se que o gás seja composto de moléculas de dois níveis com as quais o campo externo é quase ressonante. Supomos mecanismos de alargamento homogêneos (colisões duras, decaimento espontâneo) e inhomogêneas (efeito Doppler) agindo sobre o Índice de refração real do meio. A dispersão não-linear do meio é estudada como uma função das frequências de "detuning", das condições de saturação e para várias razões entre as larguras de linha homogêneas e inhomogêneas. Em particular, são enfatizadas as modificações do índice de refração devidas a efeitos de saturação.

1. INTRODUCTION

Since the discovery of anomalous dispersion by Le Roux in 1862, a great deal of research has been done on the subject. All the work done till 1931 was revised by Korff and Breit¹ and an extensive review article on the quantum theory of dispersion by Breit appeared by the same time^{2,3}.

The study of optical dispersion just before the advent of the laser was restricted to the linear regime⁴. In particular, for gases and vapors, the consideration were restricted to the homogeneous dispersion in which the thermal motion of the molecules were not taken into consideration. Under the influence of laser light the complex susceptibility depends on the intensity of the laser line and even gives rise to self focusing effects as pointed out for the first time by Javan and Kelley⁵. Other papers appeared since then on the dispersion of the resonant nonlinear susceptibility in potassium vapor⁶, rubidium vapor⁷, etc. The field of self-focusing and self-defocusing is today a well established one since the work of Chiao, Garmine, Townes⁸ and Kelley⁹. Lately, the generation of radiation by optical mixing in vapors first achieved by Ward and New¹⁰ brought the attention of researchers to another aspect of optical dispersion in gases and vapors. For the index matching is necessary to know the behavior of the index of refraction of the gases that compose the mixture. Our aim in this paper is to present an analysis of the behavior of the real index of refraction of gases and vapor in the region of anomalous dispersion (absorption region) taking into consideration the saturation effects of the laser light on the indexes of refraction, that give rise to self-focusing and self-defocusing phenomena, as well as the thermal motion of the molecules for any relative magnitude between the homogeneous and inhomogeneous absorption line widths. We assume a simple model for the gas. i.e., we assume that the molecules can be considered as two-level systems nearly resonant with the angular frequency ω_L on the laser lines. The molecules are assumed to undergo hard collisions and interact with the quantized electromagnetic free field in space giving rise to spontaneous emission. The processes of homogeneous broadening considered above are supposed to be statistically independent. We assume also that the laser action does

not modify the Maxwell-Boltzmann equilibrium distribution of the velocities of the molecules.

In section 2, we calculate the real part of the complex susceptibility of the gas (or vapor) system to be used in sections 3 in the analysis of nonlinear behavior of the index of refraction.

2. THE REAL PART OF THE COMPLEX NONLINEAR DOPPLER-LORENTZ ELECTRICAL SUSCEPTIBILITY OF THE GAS MEDIUM

As outlined in the section 1, we assume that the molecules are two-level systems near resonant with a monochromatic, linearly polarized laser field $\vec{E} = \hat{x} E_0 \cos(\omega_L t - \vec{k} \cdot \vec{r})$. The interaction between the laser field and the molecule is supposed to be taken in the linear dipole approximation. The light-gas interaction hamiltonian is then: $\hat{V} = -\vec{\mu} \cdot \vec{E}$, where $\vec{\mu}$ is the electric dipole moment operator of the molecule and \vec{E} is the electric field vector of the laser given above. The molecules are assumed to undergo hard collisions in which all phase information is lost after each collision. The molecules also can emit radiation spontaneously. The homogeneous relaxation time can be written accordingly by $\frac{1}{\tau} = \frac{1}{\tau_N} + \frac{1}{\tau_c}$, where τ_N is related to the spontaneous emission processes and τ_c related to the collisions. In both cases, the corresponding " τ " is the average time between 2 simultaneous collisions or spontaneous emissions. These two processes are assumed to be statistically independent and to obey a Poisson type distribution, namely

$$g(\theta) d\theta = e^{-\theta/\tau} \frac{d\theta}{\tau} \quad (1)$$

Expression (1) is the probability that a molecule, after surviving without collisions (and/or spontaneous decay) for a time θ , suffer a collision (and/or spontaneous decay) in the time interval between θ and $\theta + d\theta$.

To obtain the real part of the complex electrical susceptibility we proceed as follows. We describe the dynamical behavior of the two-level system with the vector model of Feynman, Vernon and Hellwarth¹¹.

In this formulation the Schrödinger equation for the two-level molecule can be written in the form

$$\frac{d\vec{r}}{dt} = \vec{\omega} \times \vec{r} \quad (2)$$

where \vec{r} describes the quantum-mechanical state of the system under the perturbation $\vec{\omega}$. For a two-level system of energies E_a and E_b ($E_a > E_b$), such that $E_a - E_b = \hbar\omega_0$ the wave-function $\psi(\vec{r}, t) = a\phi_a + b\phi_b$, where ϕ_a and ϕ_b are the eigenstates of the non-perturbed hamiltonian H_0 ,

$$\begin{cases} H_0 \phi_a = E_a \phi_a \\ H_0 \phi_b = E_b \phi_b \end{cases}$$

The components of the vector \vec{r} are defined by:

$$\begin{cases} r_1 = ab^* + ba^* \\ r_2 = i(ab^* - a^*b) \\ r_3 = aa^* - bb^* \end{cases}$$

where (*) always indicates complex conjugate. In our case, since the perturbed hamiltonian $\hat{V}(t)$ is given by

$$\hat{V}(t) = -\vec{\mu} \cdot \vec{E} = -\vec{\mu} \cdot \hat{x} E_0 \cos(\omega_L t - \vec{k} \cdot \vec{r}),$$

the vector $\vec{\omega}$ is given by its components as:

$$\begin{cases} \omega_1 = \frac{1}{\hbar} (V_{12} + V_{21}) = -\frac{\mu_{12}}{\hbar} E_0 \cos(\omega_L t - \phi_0) \end{cases} \quad (3a)$$

$$\begin{cases} \omega_2 = \frac{i}{\hbar} (V_{12} - V_{21}) = 0 \end{cases} \quad (3b)$$

$$\begin{cases} \omega_3 = \omega_0 + \frac{V_{22} - V_{11}}{\hbar} = \omega_0 \end{cases} \quad (3c)$$

where ω_0 is the resonance frequency of the two-level system, $V_{ij} = \langle i | V | j \rangle$ is the matrix element of the perturbation between the states $|i\rangle$ and $|j\rangle$ of the two-level system, $\omega_L = \omega_L (1 - \frac{v \cdot \hat{k}}{c})$ is the Doppler shifted la-

ser frequency along the direction \hat{z} of the beam propagation, ϕ_0 is a phase, and $\mu_{12} = \langle 1 | \mu | 2 \rangle$ is a matrix element of the dipole operator.

In order to solve for \vec{r} in equation (2) under the perturbation (3), we change to a rotating coordinate system M that rotates with frequency ω_- around the axis 3 in the \vec{r} space. The rotation vector is given by $\omega_- = (0, 0, \omega_-)$ and the rotation matrix is:

$$R = \begin{bmatrix} \cos \omega_- t & \sin \omega_- t & 0 \\ -\sin \omega_- t & \cos \omega_- t & 0 \\ 0 & 0 & 1 \end{bmatrix}$$

The rotated components of the vector \vec{w} are given in the well-known rotating wave approximation (RWA) by

$$\omega_1' = - \frac{\mu_{12} E_0}{\hbar} \cos \phi_0$$

$$\omega_2' = + \frac{\mu_{12} E_0}{\hbar} \sin \phi_0$$

$$\omega_3' = \omega_0$$

and the equation of motion (1) in the rotating frame by

$$\frac{dr_1'}{dt} = (\omega_- - \omega_0) r_2' + \omega_2' r_3' \quad (4a)$$

$$\frac{dr_2'}{dt} = -(\omega_- - \omega_0) r_1' + r_3' \cdot \omega_1' \quad (4b)$$

$$\frac{dr_3'}{dt} = \omega_1' r_2' - \omega_2' r_1' \quad (4c)$$

where the prime indicates the rotated quantities. Solving this system of equations assuming that the system is in its lowest energy states, i.e., $r_1'(\theta=0) = r_2'(\theta=0) = 0$, $r_3'(\theta=0) = -1$, where $\theta = t - t'$, and t' is the time of the last collision or spontaneous decay, we obtain:

$$r_1'(\theta) = \frac{\omega_1'(\omega_- - \omega_0)(1 - \cos \Omega\theta)}{\Omega^2} - \frac{\omega_2'}{\Omega} \sin \Omega\theta \quad (5a)$$

$$r_2'(\theta) = \frac{\omega_2'(\omega_- - \omega_0)(1 - \cos \Omega\theta)}{\Omega^2} + \frac{\omega_1'}{\Omega} \sin \Omega\theta \quad (5b)$$

$$r_1'(\theta) = -1 + \frac{\mu_{12}^2 E_0^2}{\hbar^2 \Omega^2} (1 - \cos \Omega\theta) \quad (5c)$$

With the Poisson distribution (1), we may consider the homogeneous average of the expectation value of the electric dipole moment operator μ induced by a uniform plane electric field wave

$$\langle \vec{\mu}(\vec{r}, t) \rangle_H = \int_0^\infty \mu_{12} r_1'(\theta) e^{-\theta/\tau} \frac{d\theta}{\tau} \quad (6)$$

from which the component of the polarization charge density \vec{P}_H corresponding to a certain package of molecules with velocity $v \approx \vec{v} \cdot \hat{z}$ can be obtained by

$$\vec{P}_H(\vec{r}, t) = N \langle \vec{\mu}(\vec{r}, t) \rangle_H \quad (7)$$

where N is the particle density.

By the other side, the homogeneous complex electrical susceptibility $\overleftrightarrow{\chi}_H(v)$ related to a package of molecules with velocity v is given by

$$\vec{P}_H(\vec{r}, t) = \text{Re} [\overleftrightarrow{\chi}_H(v) \vec{E}(\vec{r}, t)] \quad (8)$$

Using (5), (6), (7) and (8), we obtain the real part of the complex electrical susceptibility $\chi_H^1(v)$ as

$$\chi_H^1 = - \frac{N \mu_{12}^2}{\hbar} \frac{\tau^2 (\omega_- - \omega_0)}{1 + \tau^2 (\omega_- - \omega_0)^2 + \frac{\mu_{12}^2 E_0^2}{\hbar^2} \tau^2} \quad (9)$$

The imaginary part $\chi_H^2(v)$ related to the absorption of the laser wave in the medium was studied in another publication¹² and does not concern us here. We notice that in eq. (9) appear the saturation parameter $s = \frac{\mu_{12}^2 E_0^2}{\hbar^2} \tau$ that gives to the so called power broadening of the spectral lines. In the present work is the parameter that describes the

nonlinear effect of the laser field in the real index of refraction of the gas medium. To obtain the complex susceptibility of the gas, we average $\chi_H^{++}(v)$ over all velocities using the Maxwell-Boltzmann (or gaussian) distribution $f_G(\delta) = \tau_G/\sqrt{\pi} e^{-\tau_G^2 \delta^2}$ where $\delta = \omega_L/c - v$. Seen from the laboratory frame the ensemble of two-level molecules appears with resonance frequencies $\omega_0 + kv$, where $k = \omega_L/c$. The parameter τ_G is called the gaussian time and is related to the full line width $\Delta\omega_G$ at half of maximum strength of the gaussian line by

$$\Delta\omega_G = \frac{2}{\tau_G} \sqrt{\ln 2}$$

So, the real part of the inhomogeneous complex susceptibility becomes:

$$\chi_G'(\omega_L, \omega_0) = \int_{-\infty}^{+\infty} \chi_H'(\omega_L, \omega_0 + kv) \frac{e^{-\tau_G^2 \delta^2}}{\sqrt{\pi}} \tau_G d\delta \quad (10)$$

that gives

$$\chi_G'(\omega_L, \omega_0) = -\frac{N \mu_{12}^2}{\hbar} \sqrt{\pi} \tau_G \operatorname{Im} [W(z)] \quad (11)$$

where $W(z) = (e^{-z^2} \operatorname{erfc}(-iz))$ is the error function of complex argument¹³. The argument $z = x + iy$ is given by:

$$x = \tau_G(\omega_L - \omega_0) \quad (12)$$

$$y = \frac{\tau_G}{\tau} \sqrt{1 + s^2}$$

The parameter x is the detuning frequency in adimensional form and y is related to the ratio between the saturated homogeneous linewidth $\Delta\omega_H$ and the gaussian linewidth $\Delta\omega_G$:

$$y = \frac{\Delta\omega_H}{\Delta\omega_G} \sqrt{\ln 2} \quad (13a)$$

$$\Delta\omega_H = \frac{2}{\tau} \sqrt{1 + s^2} \quad (13b)$$

3. INDEX OF REFRACTION OF A GAS MEDIUM OF TWO-LEVEL SYSTEMS INTERACTING WITH A MONOCHROMATIC ELECTROMAGNETIC WAVE NEAR RESONANCE

In this section, we consider the behavior of the real part of the Doppler-Lorentz complex susceptibility $\chi_G^1(\omega_L, \omega_0)$ (ref.11) with the parameter of the model. In particular, we consider the variations of the index of refraction of the saturated gas that give rise to self-focusing and self-defocusing of the laser beam as a consequence of saturation effects.

In the simple model provided by this work for the variations for the index of refraction with the detuning frequency x , with the saturation conditions defined by the saturation parameter s , and the relationship between the homogeneous relaxation processes (collisions, spontaneous emission, etc.) and the effect of the thermal motion of the molecules, the molecule or atom is treated as a two-level system interacting with a single-frequency, plane optical wave $\vec{E} = E_0 \hat{x} \cos(\omega_L t - \phi_0)$ in the dipole wave approximation. The amplitudes of the optical wave is supposed to change negligibly between collisions and $\text{Im} k(z) \ll \text{Re} k(z)$ where $k(z)$ is the complex wave vector. The real part of the susceptibility $\chi_G^1(\omega_L, \omega_0)$ of the gas is exact in the sense that it is valid to all orders in perturbation theory. The function $\chi_G^1(\omega_L, \omega_0)$ depends on two parameters x and y . Using the tables of Abrarnovich and Stegun¹³, we plot it in Fig. (1) and Fig. (2) versus x and y , respectively. The curves in Fig. (1) are antisymmetric with respect to the origin of the coordinate axis

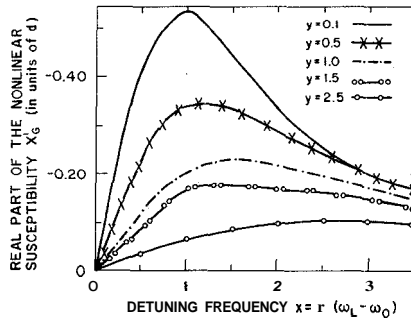


Fig.1 - Real part of the nonlinear Gaussian-Lorentzian convoluted susceptibility $\chi_G^1(\omega_L, \omega_0)$ vs Detuning frequency $x = r = \tau_G(\omega_L - \omega_0)$ for various values of the parameters $y = \tau_G/\tau \cdot \sqrt{1 + s^2}$; $d = N\mu_{12}^2/\hbar \sqrt{\pi} \tau_G$.

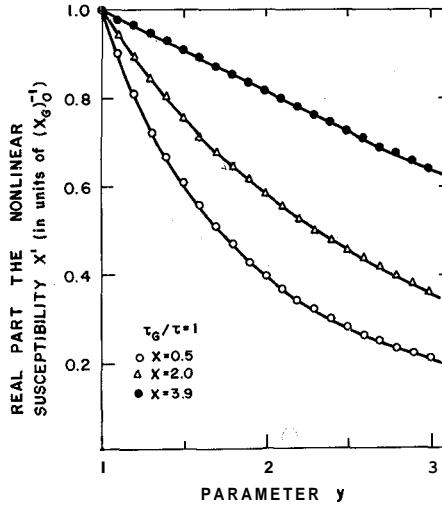


Fig.2 - Real part of the nonlinear Gaussian-Lorentzian convoluted susceptibility $\chi_G'(\omega_L, \omega_0)$ vs parameter $y = \tau_G/\tau \sqrt{1 + \varepsilon^2}$ for various values of the detuning frequency $x = \tau_G(\omega_L - \omega_0)$; $(\chi_G')_0 = \chi_G'(x, \varepsilon = 0, \tau_G/\tau)$; $\tau_G/\tau = 1$.

$$\chi_G'(x, y) = -\chi_G'(-x, y)$$

since

$$\text{Im } W(x, y) = -\text{Im } W(-x, y)$$

If the gas medium is saturated by the electromagnetic wave, its complex susceptibility becomes dependent on the strength of the field. This dependence in the imaginary part of the susceptibility is responsible for the non-exponential attenuation of the wave as it propagates through the gas, as we studied it elsewhere¹⁴. The dependence of real part of the gas susceptibility is connected with the dependence of the real index of refraction n with the external field.

The complex index of refraction n_c is related to the complex susceptibility χ_G by the expression

$$n_c^2 = 1 + 4\pi \chi_G \quad (14)$$

in C.G.S. units.

We assume, for simplicity, that the real part of the complex index variation is

$$\delta n = 2\pi \chi_G^1(\omega_L, \omega_0) \quad (15)$$

This is valid for dilute gases, and can be assumed provided the condition $\frac{4\pi}{3} |\chi| \ll 1$ holds as shown by Klein¹⁵. Of course, we could work the case in which this approximation is not valid since we have the complex susceptibility χ_G of the gas of two-level molecules.

As the electromagnetic wave propagates through the gas it is absorbed and its amplitude changes. This causes a variation of the index of refraction of the absorbing medium along the beam path.

The variation of the real index of refraction n from its field independent value n_0 is

$$n - n_0 = 2\pi(\chi_G^1 - (\chi_G^1)_0) \quad (16)$$

where $(\chi_G^1)_0$ is the field independent real part of the susceptibility. These variations of the index of refraction can then cause self-focusing or self

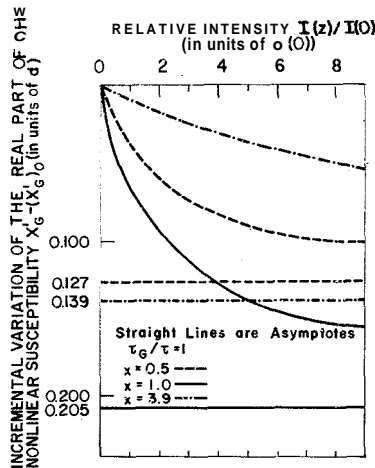


Fig.3 - Incremental variation of the real part of the nonlinear Gaussian-Lorentzian convoluted susceptibility $\chi_G^1 - (\chi_G^1)_0$ (in units of d) vs. Relative Intensity $I(z)/I(0)$ (with $\sigma(0) = (\mu_{12}^2 \tau^2 / \hbar^2) 8\pi/\sigma I(0)$) for various values of the detuning frequency $x = \tau_G(\omega_L - \omega_0)$; $d = \mu_{12}^2 / \hbar \sqrt{\pi} \tau_G$; $(\chi_G^1)_0 = x' \chi_G^1(\omega, s=0, \tau_G/\tau)$; $\tau_G/\tau = i$.

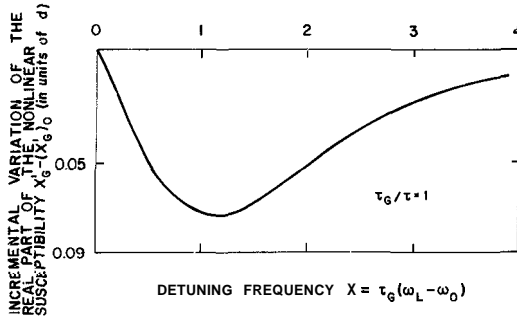


Fig.4 - Incremental variation of the real part of the nonlinear Gaussian-Lorentzian convoluted susceptibility $\chi_G^i - (\chi_G^i)_0$ (in units of d) vs. Detuning frequency $x = \tau_G(\omega_L - \omega_0)$; $r_G \hbar = 1$; $s = 1.1$; $(\chi_G^i)_0 = \chi_G^i(s=0)$; $d = \mu_{12}^2 / \hbar \sqrt{\pi} \tau_G$; $s = \mu_{12} E_0 / \hbar$ $r =$ Saturation Parameter.

-defocusing of the laser beam. In Fig. (3), the variation of the index of refraction of the medium for different saturation conditions on different points in the beam path in the gas is shown. The plot is made for several detuning frequency differences.

We notice that for $x > 0$ ($\omega_L > \omega_0$) the difference $\chi_G^i - (\chi_G^i)_0$ is positive implying $n > n_0$, i.e., focusing of the beam. This focusing effect varies with the detuning frequency and reaches a maximum for $x \approx 1.1$. This is clearly seen in Fig. (4). In Fig. (5) several curves are drawn for different values of the parameter y , i.e., for different saturation parameters and different thermodynamic (temperature, pressure, etc.) conditions.

The function $\chi_G^i - (\chi_G^i)_0$ is an odd function of x . This leads to the maximum defocusing being symmetric with respect to x to the maximum focusing. If the medium amplifies the radiation instead of absorbing it, the situation reverses, Fig. (5) and Fig. (6) display the incremental ratio $\chi_G^i - (\chi_G^i)_0 / (\chi_G^i)_0$ as a function of s^2 and x respectively.

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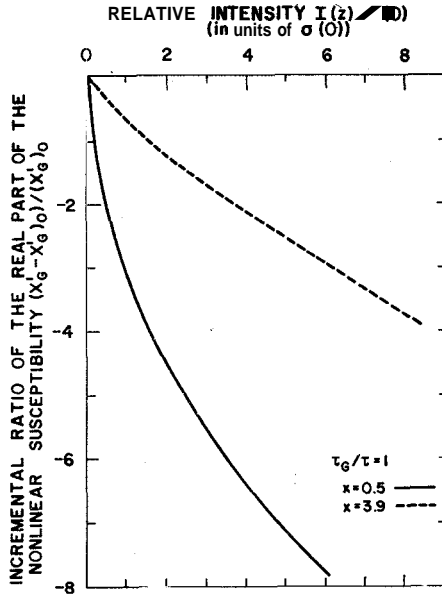


Fig.5 - incremental Ratio of the real part of the nonlinear Gaussian-Lorentzian convoluted susceptibility vs. Relative Intensity $I(z)/I(0)$ (in units of $|\sigma(0)|^{-1}$) of a strong monochromatic EM wave ($\epsilon = 2.8$) for various values of the detuning frequency $x = \tau_G(\omega_L - \omega_0)$; $\tau_G/\tau = 1$; $\sigma(0) = s^2(0) = \mu_{12}^2 \tau^2 / \pi^2 \cdot 8\pi/c I(0)$; $(\chi'_G)_0 = \chi'_G(s=0)$; Saturation Parameter $s = \mu_{12} E_c / \hbar \tau$.

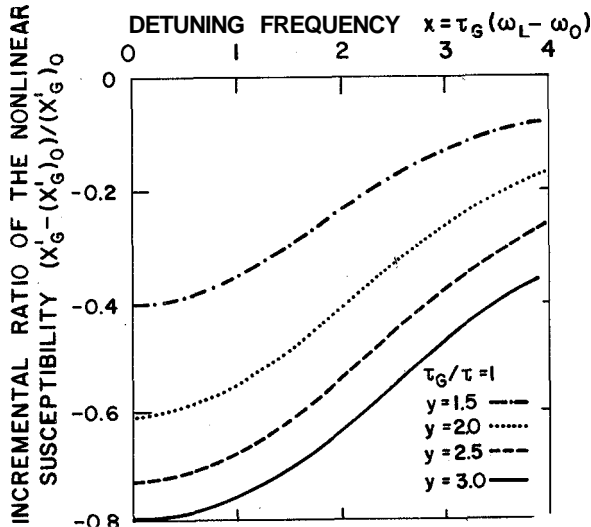


Fig.6 - Incremental Ratio of the real part of the nonlinear Gaussian-Lorentzian convoluted susceptibility vs. Detuning Frequency $x = \tau_G(\omega_L - \omega_0)$ for various values of the parameter $y = \tau_G/\tau \sqrt{1 + s^2}$; $\tau_G/\tau = 1$.

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