Limiting efficiencies of parametric frequency converters with optically inhomogeneous nonlinear media

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(Submitted July 11, 1975)
Zh. Eksp. Teor. Fiz. **70**, 458–466 (February 1976)

A method is developed for obtaining steady-state solutions of reduced equations for the nonlinear interaction of waves in inhomogeneous media without recourse to the approximation of a given pump field. The method is used in an analysis of the efficiency of optical frequency doublers under conditions of regular or random loss of phase matching. It is shown that the 100% efficiency limit is unattainable if such loss of phase matching does occur and in this case the efficiency initially rises with pump power, reaches a certain maximum value for a crystal of length L, and then begins to fall. A full analytic treatment of the efficiency of a frequency doubler in the presence of accidental loss of phase matching makes it possible to calculate the optimal length of a crystal L_{max} as a function of the pump power for which the doubler efficiency is maximal η_{max}. It is shown that the limiting efficiency η_{max} of a crystal with the optimal length L_{max} (L_{max} decreases with rising power) rises with the pump power until the thermal self-interaction and diffraction-caused loss of phase matching become important.

PACS numbers: 42.65.−k, 42.20.−y

1. INTRODUCTION

In most cases of interest the nonlinear interaction of waves is accompanied by regular or random loss of phase matching of the waves because of various factors, such as the inhomogeneity of the medium, diffraction, thermal self-interaction, etc. This destroys the phase matching to which the resonant interactions of the waves are sensitive and the general picture of nonlinear wave processes is found to depend strongly on such factors. Resonant nonlinear interactions involving a small number of waves underlie the operation of parametric converters of the frequency of light. The recent developments in the technology of stable single-mode lasers and the possibility of overcoming aperture effect have raised in acute form the problem of achieving the maximum possible efficiencies in optical frequency doublers and optical parametric oscillators.[1–3]

The most important factor which limits the efficiency of parametric frequency converters is the loss of phase matching and the consequent weakening of the nonlinear interaction.[1,4] For example, computer calculations have shown that the theoretical efficiency of 100% is unattainable in optical frequency doublers for finite beams because of the diffraction-induced loss of phase matching.[4,5] A consistent analytic discussion of the limiting efficiencies of parametric frequency converters in the presence of regular or random loss of phase matching requires avoidance of the approximation of a given pump field, which is usually employed (see, for example,[6,7]) and reduces essentially the problem to a linear one. The difficulties encountered outside the framework of a given field approximation are the strong nonlinearity of the problem and the absence of general solution methods.

We shall develop a method for obtaining steady-state ($\partial/\partial t = 0$) solutions of one-dimensional (all quantities depend on just one coordinate) reduced equations for the nonlinear interaction of three waves in inhomogeneous media used earlier[8,9] in discussing the nonlinear interaction of waves in a nonequilibrium inhomogeneous plasma. The approximation of a given field is not invoked and the amplitudes of all three waves can be of the same order of magnitude. The small parameter used in the present paper is related to the smallness of the accumulated relative difference between the wave phases due to the inhomogeneity of the medium in a characteristic nonlinear interaction length l. This parameter is perfectly natural in the problem of limiting efficiencies of parametric converters because if the accumulated difference between the wave phases in a distance l is greater than, or of the order of, unity, the nonlinear interaction becomes much weaker and we cannot expect high efficiencies.

The method will be used to analyze second harmonic generation in a medium with a regular or random loss of phase matching because of inhomogeneities. It will be shown that, for a fixed length of a nonlinear medium the efficiency of second harmonic generation does not saturate at the limit of 100% when the power is increased, which could be expected in the absence of any change in the phase matching in a lossless medium, but it begins to fall from limiting value lower than 100% and the energy is then pumped back to the first harmonic. Similar behavior of the efficiency of an optical doubler, represented by the dependence of the total power on the reduced length of a nonlinear crystal, is reported by Karamzin and Sukhorukov.[4] An analysis shows that the cause of this phenomenon is the spatial development of an instability of the second harmonic which decays into two waves of frequency ω and in this case the loss of phase matching acts as a perturbation nucleus. This instability of the maximum-frequency wave leading to a decay into two waves with lower frequencies has been studied thoroughly in the theory of plasma and is known as the decay instability.[10]

The solution obtained will enable us to determine the theoretical efficiency limit of a frequency doubler in the presence of accidental loss of phase matching between the waves, and to find the optimal conditions for the operation of a doubler. Thus, if the parameters representing the accidental loss of phase matching are fixed, we find that—for a given length of a nonlinear crystal—there is an optimal pump power at which the efficiency reaches its maximum.

2. SOLUTION METHOD

The steady-state ($\partial/\partial t = 0$) process of second harmonic generation in a weakly nonlinear ($kl \gg 1$) weakly inhomogeneous ($kl_c \gg 1$) dissipative medium is described by the following system of equations for the complex amplitudes of the first and second harmonics $A_{1,2}(z)$ (the z axis is perpendicular to the boundary of the nonlinear medium[7,11]:

$$\frac{dA_1}{dz}+b_1A_1=i\beta_1A_2A_1^*+i\Lambda_1(z)A_1,$$
$$\frac{dA_2}{dz}+b_2A_2=i\beta_2A_1^2+i\Lambda_2(z)A_2. \quad (1)$$

Here, l_c is the characteristic scale of inhomogeneities, $b_{1,2}$ are the damping decrements of the harmonics, $\beta_{1,2}$ are the nonlinear constant interaction coefficients governed by the frequencies and wave vectors of the waves, and by the nonlinear susceptibility tensor of the medium (see, for example, [11]).

The phase matching conditions are assumed to be satisfied by the waves at $z = 0$ and the loss of phase matching between the waves in the medium is related to the factors $\Lambda_{1,2}(z)$ [$\Lambda_{1,2}(0) = 0$]. If the loss of phase matching is due to the inhomogeneity of the medium, we have

$$\Lambda_1(z)=\frac{\omega^2}{c^2k_1\varepsilon_1}\frac{\delta\varepsilon_1(z)}{\varepsilon_1},\qquad \Lambda_2(z)=\frac{(2\omega)^2}{c^2k_2\varepsilon_2}\frac{\delta\varepsilon_2(z)}{\varepsilon_2}, \quad (2)$$

where ω is the frequency of the first harmonic, $k_{1,2}$ are the projections of the wave vectors onto the z axis, $\epsilon_{1,2}$ and $\delta\epsilon_{1,2}(z)$ are the constant and alternating parts of the permittivity. The factors $\Lambda_{1,2}(z)$ are clearly related as follows to that part of the phases $\Phi_{1,2}(z)$ of the complex wave amplitudes $A_{1,2}(z)$ which changes due to external factors (i.e., $b_{1,2} = \beta_{1,2} = 0$):

$$\Lambda_i(z)=\frac{d\Phi_i(z)}{dz},\qquad i=1,2. \quad (3)$$

Let $\rho_i(z)$ and $\varphi_i(z)$ be the moduli and phases of the complex amplitudes of the waves $A_i(z) = \rho_i(z)\exp[i\varphi_i(z)]$. Then, it is convenient to use the following real dimensionless quantities:

$$n_1(z)=\frac{\rho_1^2(z)}{\rho_1^2(0)},\qquad n(z)=\frac{\rho_2^2(z)}{\rho_2^2(0)}\frac{\beta_1}{\beta_2},$$
$$x=z/l,\quad l^{-1}=\beta_1(\beta_2)^{1/2}\rho_1(0),$$
$$\theta=\varphi_2(z)-2\varphi_1(z). \quad (4)$$

We shall adopt the following boundary conditions: $n_1(0) = 1$, $n(0) = 0$, $\theta(0) = \pi/2$. In terms of these variables and using the Manley–Rowe relationship $n_1(x) + n(x) = 1$ in a lossless medium ($b_1 = b_2 = 0$), the system (1) reduces to two equations for the normalized intensity of the second harmonic $n(x)$ and the relative difference between the phases of the waves $\theta(x)$:

$$\frac{dn}{dx}=2n^{1/2}(1-n)\sin\theta, \quad (5)$$

$$\frac{d\theta}{dx}=\varkappa(x)+\left(\frac{1-n}{n^{1/2}}-2\sqrt{n}\right)\cos\theta, \quad (6)$$

$$\varkappa(x)=l\Delta k(lx),\quad \Delta k(x)=\Lambda_2(lx)-2\Lambda_1(lx). \quad (7)$$

The system (5) satisfies the following integral relationship:

$$n^{1/2}(1-n)\cos\theta+\frac{1}{2}\int_0^x dx_1\,\varkappa(x_1)\frac{dn}{dx_1}=\Gamma. \quad (8)$$

The quantity Γ is governed by the boundary conditions. If $\varkappa(x) = \text{const}$, this integral relationship reduces to the well-known integral of motion of the system (5)–(6).[11] We shall express $\cos\theta$ in terms of $n(x)$ using Eq. (8) and we shall substitute the resultant expression in Eq. (6). We shall then integrate formally the left- and right-hand sides of Eq. (6). Then, we shall substitute the expression for the phase $\theta(x)$ into Eq. (5) and obtain finally one integro-differential equation for $n(x)$ (a similar equation was used earlier in[8,9]):

$$\frac{dn}{dx}=2n^{1/2}(1-n)\sin\left\{\frac{\pi}{2}+\int_0^x dx_1\,\varkappa(x_1)+\int_0^x dx_1\left[\frac{1}{1-n(x_1)}-\frac{1}{2n(x_1)}\right]\int_0^{x_1}dx_2\,\varkappa(x_2)\frac{dn}{dx_2}\right\}. \quad (9)$$

In Eq. (9) the initial phase shift between the waves is $\theta(0) = \pi/2$ ($\Gamma = 0$).

We shall introduce

$$\alpha\{n(x)\}=\int_0^x dx_1\,\varkappa(x_1)+\int_0^x dx_1\left[\frac{1}{1-n(x_1)}-\frac{1}{2n(x_1)}\right]\int_0^{x_1}dx_2\,\varkappa(x_2)\frac{dn}{dx_2}; \quad (10)$$

where $\alpha\{n(x)\}$ represents the accumulation of the phase shift (because of the inhomogeneity of the medium) in a distance l. It follows qualitatively from Eq. (9) that if the accumulated phase shift is $\alpha\{n(x)\} \gtrsim 1$, the sine in Eq. (9) begins to oscillate and the effective interaction between the waves weakens considerably. We shall therefore assume that the influence of the loss of phase matching on the nonlinear interaction of the waves is weak, so that the quantity $\alpha\{n(x)\}$ can be assumed to be small and the solution of Eq. (9) will be obtained as a series in perturbation theory. The final solution for the normalized intensity of the second harmonic $n(x)$ including the first term of the perturbation series is

$$n(x)=\tanh^2(x)\left[1-\frac{1}{2\,\mathrm{sh}\,2x}\int_0^x dx_1\,\alpha^2(x_1)\right]. \quad (11)$$

Here, $\alpha(x)$ is given by Eq. (10), where $n(x)$ should be replaced with the unperturbed solution $n_0(x) = \tanh^2(x)$. The criterion of the validity of the solution (11) is the closeness of the solution to the unperturbed form, i.e., the smallness of the value

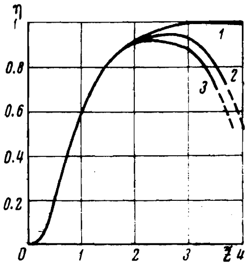

FIG. 1. Dependence of the doubler efficiency on the reduced length of the nonlinear crystal. Curve 1 corresponds to $AL=0$, curve 2 to $AL=0.05$, and curve 3 to $AL=0.1$.

$$\frac{1}{2\,\mathrm{sh}\,2x}\int_0^x dx_1\,\alpha^2(x_1)\ll 1. \tag{11a}$$

The expression (11) describes the efficiency of a frequency doubler for the wave intensities and also for the total power across the beam, because we are considering only beams with homogeneous transverse distributions and with transverse dimensions much greater than l. It is clear from Eq. (11) that if the loss of phase matching occurs, $\varkappa(x)\neq 0$, the efficiency $\eta=n(x)$ of a doubler is always less than unity.

3. SECOND HARMONIC GENERATION IN A MEDIUM WITH RANDOM INHOMOGENEITIES

We shall consider the case when the loss of phase matching is random and the properties of the quantity $\varkappa(x)$ are known (Bespalov[12] solved the problem of frequency doubling in the presence of random loss of phase matching but he used the approximation of a given pump field and somewhat different assumptions about the nature of the random process). We shall assume that the nonlinear interaction coefficients are constant and that the refractive index of the medium is a fluctuating quality, which corresponds to the case of optically inhomogeneous nonlinear crystals.

Let $\varkappa(x)$ be a steady-state Gaussian random process with zero mean $\langle\varkappa(x)\rangle=0$ and pair correlation function $B(x_1-x_2)=\langle\varkappa(x_1),\varkappa(x_2)\rangle$. Then, the solution of the problem of the doubler efficiency $\eta=\langle n(x)\rangle$ reduces to the averaging over the random process in Eq. (11) and subsequent calculation of the integrals

$$\langle n(x)\rangle=\tanh^2(x)\left[1-\frac{1}{2\,\mathrm{sh}\,2x}\int_0^x dx_1\langle\alpha^2(x_1)\rangle\right]. \tag{12}$$

We shall consider the case of short correlation lengths l_c ($\gamma^{-1}\equiv l_c/l\gg 1$). In this case we can assume that in the averaging process we have

$$B(x_1-x_2)=B\delta(x_1-x_2). \tag{13}$$

This approximation corresponds to the retention of only the first terms in respect of the parameter $\gamma^{-1}\ll 1$. The higher terms of the expansion in γ^{-1} can be found using, for example, a correlation function of the type

$$B(x_1-x_2)=\langle\varkappa^2\rangle\exp(-\gamma|x_1-x_2|), \tag{14}$$

where $\langle\varkappa^2\rangle=\langle\Delta k^2\rangle l^2$ is the mean square of the phase shift accumulated in a distance l.

Retention of only the first term in γ^{-1} corresponds formally to going to the following limit in Eq. (14):

$$\gamma^{-1}\to 0,\quad \langle\varkappa^2\rangle\to\infty,\quad 2\langle\varkappa^2\rangle\gamma^{-1}\to B=\mathrm{const},$$
$$B(x_1-x_2)\to B\delta(x_1-x_2). \tag{15}$$

This establishes the meaning of the quantity B:

$$B=2\langle\Delta k^2\rangle l l_c\equiv Al,\quad A=2l_c\langle\Delta k^2\rangle. \tag{16}$$

The quantity A represents the properties of the medium. Averaging $n(x)$ in Eq. (12) over the random process characterized by the correlation function (13) and calculating the integrals, we obtain the following expression for the doubler frequency:

$$\eta(x)\equiv\langle n(x)\rangle=\tanh^2(x)\left[1-B\frac{\mathrm{ch}\,4x+16\,\mathrm{ch}\,2x-17}{120\,\mathrm{sh}\,2x}\right]. \tag{17}$$

We shall also give the formula for the doubler efficiency which is more convenient in the case when the length L of a crystal is fixed and only the incident wave power, i.e., the length l, is varied:

$$\eta(\tilde z)\equiv\langle n(\tilde z)\rangle=\tanh^2(\tilde z)\left[1-\frac{AL}{120\tilde z}\left(\frac{\mathrm{ch}\,4\tilde z+16\,\mathrm{ch}\,2\tilde z-17}{\mathrm{sh}\,2\tilde z}\right)\right], \tag{17a}$$

where $\tilde z=L/l$ is the reduced length of the nonlinear crystal. The dependences $\eta(\tilde z)$ calculated for different values of AL are plotted in Fig. 1.

The pumping of the energy back to the first harmonic is described by

$$\langle n_1(x)\rangle=1-\langle n(x)\rangle. \tag{18}$$

The expressions (17) and (18) show that the pumping of the energy back to the first harmonic due to the phase shift occurs in a characteristic distance z_s:

$$z_s\sim l\ln(120B^{-1})^{1/2}. \tag{19}$$

The physical cause of this rise of the first harmonic intensity is the spatial growth of the decay instability of a wave frequency 2ω into two waves of frequencies ω,[10,11] which in this case is of fluctuation nature. The length z_s is also the distance in which the fluctuation component $\tilde n(x)$ becomes comparable in order of magnitude with $\langle n(x)\rangle$ and we can no longer use the approximation described above [the small parameter $e^x(B/120)^{1/2}\ll 1$ becomes strongly dependent on the length].

We can quote quantitative considerations to support that the characteristic length z_s of the process of pumping the energy back to the first harmonic is a logarithmic function of the perturbation, irrespective of its actual nature. In fact, let the ratio of the intensity of a perturbation at a frequency ω to the intensity of the second harmonic be of the order of B. Then, it is well known that the characteristic length of the decay of wave frequency 2ω is (see, for example,[10] p. 22)

$$z_s\sim{}^1/_2 l\ln B^{-1},\quad B\ll 1. \tag{20}$$

We shall conclude this paragraph by noting that the problem of three-wave interaction in a randomly homogeneous medium with special boundary conditions, solved by one of the present authors by a different method,[13] gives similar results and a characteristic length of increase of fluctuations of the same type as Eq. (20). The approximation employed by Tamoĭkin and Faĭnshteĭn[14] in an investigation of three-wave processes in random media corresponds to dropping of the second term from the formula (10) for $\alpha\{n(x)\}$.

4. OPTIMAL PARAMETERS OF A FREQUENCY DOUBLER IN THE CASE OF RANDOM LOSS OF PHASE MATCHING

We shall now solve the problem of optimal parameters of a doubler in the case when the main efficiency-limiting factor is a random inhomogeneity in the nonlinear crystal. For convenience, we shall assume that the properties of this crystal and the incident power are constant, i. e., that $B = Al$ is a fixed quantity, but the length of the crystal L can be varied. The final formulas are not affected if AL is regarded as fixed and l is varied. The solution (17) shows that, for a fixed value of B, there is an optimal dimensionless length of a nonlinear crystal $x_{\max} = L_{\max}/l$ in which the doubler efficiency reaches its maximum value $\eta_{\max}$. Further increase of x causes the doubler efficiency to fall.

The dependence of $x_{\max}$ on the parameter B is governed by the following transcendental equation:

$$\left.\frac{d\eta(x)}{dx}\right|_{x=x_{\max}}=0, \tag{21}$$

where $\eta(x)$ is given by Eq. (17). We shall solve this equation on the assumption of sufficiently low values of B, so that $x_{\max} \gtrsim 2$. Then, the hyperbolic functions in Eq. (21) can be replaced with exponential functions and Eq. (21) becomes

$$\exp(4x_{\max})=480B^{-1},\quad L_{\max}/l\equiv x_{\max}={}^1/_4\ln[480B^{-1}]. \tag{22}$$

Substituting Eq. (22) into Eq. (17), we obtain the dependence of the maximum efficiency of second harmonic generation $\eta_{\max}$ on the parameter B in the presence of random loss of phase matching:

$$\eta_{\max}\approx 1-2(B/30)^{1/2}\approx 1-0.4(Al)^{1/2}. \tag{23}$$

It should be pointed out that the validity of the above approximations in the region $x_{\max}$ corresponding to the efficiency maximum is subject to the inequality $0.4B^{1/2} \ll 1$.

We shall now consider the results obtained, for example those for the loss of phase matching due to a random inhomogeneity. Then, in Eq. (16) the expression for $B = 2\langle\Delta k^2\rangle l l_c$ should be modified for order-of-magnitude estimates by replacing $\langle\Delta k^2\rangle$ with [see Eq. (2)]

$$\langle\Delta k^2\rangle\sim k^2\langle\delta\varepsilon^2\rangle/\varepsilon^2, \tag{24}$$

where $\langle\delta\varepsilon^2\rangle$ is the mean square of the spatial fluctuations of the permittivity: $B \sim k^2 l l_c \langle\delta\varepsilon^2\rangle/\varepsilon^2$. If l_c and $\langle\delta\varepsilon^2\rangle/\varepsilon^2$ are fixed ($A = \text{const}$), an increase in the incident power causes the maximum efficiency $\eta_{\max}$ to rise, in accordance with Eq. (23), because l decreases. The optimal length of the nonlinear crystal

$$L_{\max}=\frac{l}{4}\ln\left[480\frac{\varepsilon^2}{\langle\delta\varepsilon^2\rangle}\frac{1}{kl_c l}\right], \tag{25}$$

in which the maximum efficiency is reached, decreases when the incident power is increased, whereas the dimensionless length $x_{\max} = L_{\max}/l$ becomes greater. Naturally, the above estimates cease to be valid when the power of the incident wave is so high that the thermal self-interaction effects become important.

5. POSSIBLE GENERALIZATIONS

It is interesting to consider simultaneously the influence of losses and of random dephasing on the limiting efficiency of optical doublers. We can consider analytically the case of equal damping decrements of the first and second harmonics: $b_1 = b_2 = b$. The substitution of variables[11] reduces the frequency doubling problem to that solved above subject to the factor that the correlation function is now of somewhat different form when expressed in terms of new variables. Next, considering the cases of low losses ($bl \ll 1$, $bz \ll 1$) we obtain the following expression for the doubler efficiency averaged over the random process and considered to be a function of the dimensionless length of the nonlinear crystal:

$$\eta(x)=\langle n(x)\rangle=\tanh^2(x)\left[1-2blx\left(1+\frac{x}{\operatorname{sh}2x}\right)-\frac{Al}{120}\frac{\operatorname{ch}4x+16\operatorname{ch}2x-17}{\operatorname{sh}2x}\right]. \tag{26}$$

This formula shows that when the dimensionless length of the nonlinear crystal is increased, the influence of a random inhomogeneity on the doubler efficiency rises exponentially and the relative influence of the losses on the efficiency, compared with the dephasing, decreases.

The method developed in the present paper can be generalized to the case of interaction of three waves of different frequencies in inhomogeneous media. Once again use is made of one integrodifferential equation for the intensity of only one of the interacting waves and perturbation theory can be based on a small parameter representing the accumulation of the phase shift of the waves in a characteristic nonlinear interaction length, governed by the boundary conditions of the problem. In the general case of arbitrary boundary conditions the problem can be solved in the form of integrals of known functions for which analytic estimates can be obtained only in certain limiting cases. The method can be useful also in considering the nonlinear interaction of waves in an inhomogeneous plasma.[6,15,16]

6. CONCLUSIONS

An analysis of the frequency doubler efficiency shows that the limiting efficiency of 100% for ideal crystals is unattainable in the case of regular or random loss of phase matching (dephasing) of waves. In the case of a fixed length of an optically inhomogeneous nonlinear crystal L, the doubler efficiency first rises with the incident power, reaches a certain maximum value (for a given length of the crystal), and then falls when the pump power is increased still further. For a given pump power and given properties of the crystal there is an optimal length of the crystal L_{max} for which the doubler efficiency has its maximum (under these conditions) value η_{max}. Under given conditions in the nonlinear crystal the maximum possible efficiency η_{max} rises with the pump power but this value of η_{max} can be obtained only by selecting the optimal length of the crystal L_{max} which varies with the pump power. The conclusion of the rise of η_{max} with increasing pump power ceases to be valid at powers such that the thermal self-interaction effects and diffraction-induced loss of phase matching become important.

The authors are deeply grateful to S. A. Akhmanov for valuable discussions of some of the problems.

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Translated by A. Tybulewicz

Classical heteropolar molecule in the field of circularly polarized laser radiation

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(Submitted July 14, 1975)
Zh. Eksp. Teor. Fiz. 70, 467–476 (February 1976)

An approximate system of equations is obtained for the evolution of the state of a classical molecule. It is shown that the rotation of such a molecule has a strong influence on the buildup of radial vibrations: under certain conditions the rotation may compensate the radial vibration anharmonicity which disturbs the buildup. The dependence of the energy of this molecule on the radiation field intensity is considered; it is shown, in particular, that under certain conditions the rotation energy may be much greater than the vibration energy.

PACS numbers: 32.20.Tg

1. FORMULATION OF THE PROBLEM, DESCRIPTION OF THE MODEL, AND SOLUTION METHOD

We shall consider a diatomic heteropolar isolated molecule subjected to the field of monochromatic circularly polarized laser radiation. The question is what energy is transferred from the field to the molecule and how is this energy distributed between vibrational and rotational motion.

We shall consider this problem on the basis of the following model. We shall assume that the molecule consists of two point charged atoms. The force exerted on the atoms by the laser radiation is weak compared with the intramolecular force. Radial vibrations (vibrations of the distance between the atoms) are generally anharmonic but the amplitude of these vibrations is small compared with the equilibrium distance. The action of the laser radiation, rotation, and vibration of the molecule will all be considered ignoring the quantum effects.

The approximate nature of this model is self-evident. However, this model should not be considered just as