

Generalized treatment for diffusion waves

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Intended for teaching purposes, the phenomenon of diffusion in the presence of periodical sources is described, taking into account a characteristic operator, $\hat{F}(t)$, leading to a generalized hyperbolic equation. The essential features of the accompanying harmonic flux are presented. For this purpose the solution to the problem is interpreted in terms of diffusion waves, a peculiar class of waves with complex wave numbers whose generation, propagation and detection constitute the basis of modern analytical techniques able to measure optical and transport properties of materials in the condensed or gaseous phase. A generalized mathematical equation describing this kind of waves is shown and the existence of critical modulation frequencies, at which the diffusive fluxes change their behaviour, is demonstrated for different physical phenomena involving diffusion waves. The dispersion equation for diffusion waves is given, and different particular cases in modulation frequency “spectrum” are discussed.

Keywords: Diffusion; periodical sources; dispersion equation.

Con propósitos de enseñanza se describe el fenómeno de difusión en presencia de fuentes periódicas teniendo en cuenta un operador característico, $\hat{F}(t)$, que conduce a una ecuación de difusión hiperbólica generalizada. Se presentan las características fundamentales del flujo de calor armónico asociado a ella. Para ello se interpreta la solución del problema en términos de ondas de difusión, un tipo particular de ondas con números de onda complejos y cuya generación, propagación y detección constituyen las bases de técnicas analíticas modernas capaces de medir propiedades ópticas y de transporte de materiales en la fase condensada o gaseosa. Se presenta una ecuación matemática generalizada para describir esta clase de ondas y se demuestra para diferentes fenómenos que involucran las ondas de difusión la existencia de frecuencias de modulación características a las cuales el flujo difusivo cambia su carácter. Se presenta la ecuación de dispersión y se discuten diferentes casos particulares en el “espectro” de frecuencia de modulación.

Descriptores: Difusión; fuentes periódicas; ecuación de dispersión.

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1. Introduction

In the last 30 years, the concept of Diffusion Waves [1] has been increasingly used for the description of several physical phenomena [2-9] for which the presence of a periodically varying source is common. Therefore, many authors have adopted the concepts of wave physics that were used successfully in the explanation of other periodic phenomena, to interpret their experimental results.

The concept of waves is involved in many fields of science, and is becoming an integral part of the physics curricula at different levels. The increasing use of concepts related to wave propagation in the teaching of physics argues in favor of their introduction whenever possible. As the analysis of transport problems presented in standard textbooks does not make systematic use of the wave treatment, it is the purpose of this work to discuss some aspects of the so-called diffusion waves, *i.e.*, solutions of the diffusion equation in the case of periodic modulated sources. A generalized equation is presented describing diffusion wave fields of a general nature. For time-varying harmonic sources, and based on the dispersion relation, we shall emphasize some of the physical aspects related to the nature of the diffusion fields and their frequency dependence, attempting to present these questions in a unified style so that their use for educational aims will be favored.

2. Theory

Consider a sample where oscillatory wave fields $\varphi(\vec{r},t)$ are generated by a source with periodically modulated intensity, or driving force [1], of the form $Q(\vec{r})e^{i\omega t}$, where ω is the angular modulation frequency, \vec{r} is the spatial coordinate, and t is time. The function $\varphi(\vec{r},t)$ can be a thermal or temperature wave, $T(\vec{r},t)$ [10]; or a charge carrier density wave, $N(\vec{r},t)$ [11], in photothermal [3] experiments; a diffuse photon density wave in a turbid medium excited by periodically infrared light, $u(\vec{r},t)$ [2], among others. These phenomena will be used as illustrative examples for the discussion. For example, diffusion waves were used in the past in the analysis of compound migration in stratified media [4]; in the study of molecular diffusion processes by means of pressure oscillations in vacuum chambers [5]; as well as in applications related to mass transport in metals [6], electrolytes [7] and dialysis membranes [8]. Early works concerning the diffusion of a periodic flux of neutrons were also reported elsewhere [9].

We shall assume, as is often encountered in the praxis, that the sample is homogeneous and isotropic, and its properties are constant throughout the changes in temperature involved. The results achieved here can be extended by the interested reader, with suitable modifications, to a more general situation.

As mentioned in Ref. 1, the field distribution within the sample can be described by the homogeneous equation:

$$\nabla^2 \varphi(\vec{r}, t) - \frac{1}{D} \frac{\partial \varphi(\vec{r}, t)}{\partial t} - \hat{F}(t) \varphi(\vec{r}, t) = 0 \quad , \quad (1)$$

where ∇^2 is the Laplace operator, r is the spatial coordinate, t is time, $\hat{F}(t)$ is an operator characterizing a given phenomenon, and D can be interpreted as a medium characteristic transport parameter, usually a diffusion coefficient in m^2/s units. It becomes the material's thermal diffusivity, α , in the case of thermal waves, the carrier diffusion coefficient, D_n , for plasma waves in semiconductors and the light diffusion coefficient, D_l , for a diffusing photon flux in turbid media.

A brief discussion concerning the operator $\hat{F}(t)$ is introduced at this point. It is usually taken as a (real) scalar constant, say β^2 , given by the inverse of the squares of the diffusion lengths of the wave's carriers [1]: $\hat{F}=1/L_n^2$ for plasma waves in semiconductors, *i.e.* the distance a free carrier travels before it recombines with ones of opposite sign, and $\hat{F}=1/L_l^2$ for diffusing photon waves, *i.e.*, the inverse of the distance a photon will travel under random motion until it is absorbed by the medium. For thermal waves, on the other hand, it is usually assumed that $\hat{F}=0$, giving rise to instantaneous heat propagation or infinite speeds of propagation, as described by several authors [1,10,12-14]. If we consider, for example, a flat slab and a supply of heat is applied, at a given moment, to one of its faces, then according to Eq. (1), there is an instantaneous effect to the rear. The same thing occurs in the case of Fick's (first and second) Laws of diffusion, where the flux of the diffusing object (mass) reacts simultaneously to the concentration gradient leading to an unbounded propagation speed. This conclusion, of course, is physically unreasonable. If \hat{F} is a constant scalar, as in the examples given above, then Eq. (1) becomes parabolic (with a first-order derivative in time and a second-order one in the spatial coordinate), while wave equations are hyperbolic (with second-order derivatives in both space and time coordinates). This fact can be argued to assert that, according to Eq. (1), the magnitude $\varphi(\vec{r}, t)$ propagates in a non-wavelike manner, although nowadays, the theories based on a wave treatment of its propagation have been successfully used to interpret the experimental data in experiments related to the above-mentioned phenomenon.

To rectify the above-described weaknesses, one can redefine the operator \hat{F} as

$$\hat{F}(t) = \frac{1}{u^2} \frac{\partial^2}{\partial t^2} + \beta^2 \quad , \quad (2)$$

introducing the second time derivative, where u is a parameter having the dimensions of a velocity. Consequently Eq. (1) becomes hyperbolic (note its analogy with the simplest form of the well-known telegraphist equation resulting when the inductance is incorporated into Ohm's law for an electrical conductor, implying that electromagnetic signals propagate with a finite velocity, in agreement with experimental evidence). It is worth recalling that the term containing β^2 is

introduced as an *ad-hoc* condition in order to account for the discussion given above concerning the significance of the $\hat{F}(t)$ operator.

We shall, from now on, consider a source of waves located at the (semi-infinite) sample's surface, and we shall restrict our analysis to the one-dimensional case, *i.e.* we shall assume the source to be uniformly distributed across the surface. Then we have to solve the equation resulting from substituting Eq. (2) in Eq. (1), *i.e.*

$$\frac{\partial^2 \varphi(x, t)}{\partial x^2} - \frac{1}{D} \frac{\partial \varphi(x, t)}{\partial t} - \frac{1}{u^2} \frac{\partial^2 \varphi(x, t)}{\partial t^2} - \beta^2 \varphi(x, t) = 0 \quad (3)$$

Now we shall try to explain the origin of the second time derivative term on the basis of the heat transport phenomenon, where it often appears. Note that when the wave field is the temperature, and for $\beta^2=0$, Eq. (3) becomes the well-known hyperbolic heat diffusion equation [13,14]. If, as a consequence of the temperature gradient existing at each time instant, t , the heat flux appears only at a later instant, $t + \tau$, then Fourier's Law adopts the form:

$$q(x, t + \tau) = -k \frac{\partial T(x, t)}{\partial x} \quad (4)$$

Time τ is the thermal relaxation time, *i.e.* the build-up time for the onset of the thermal flux after a temperature gradient is suddenly imposed on the sample. It is associated with the phonon-phonon interaction after the start of the diffusive heat flux. If τ is small, then we can expand the heat flux in a Taylor Series around $\tau = 0$ obtaining:

$$q(x, t + \tau) = q(x, t) + \tau \frac{\partial q(x, t)}{\partial t} \quad (5)$$

where we neglect higher order terms. Substituting Eq. (5) into Eq. (4) leads to:

$$q(x, t) + \tau \frac{\partial q(x, t)}{\partial t} = -k \frac{\partial T(x, t)}{\partial x} \quad (6)$$

This is the so-called modified Fourier's law, also known as Cattaneo's Equation (we can find equations of the same kind as Eq. (4) for other wave fields, for example the first Fick's Law of Mass Diffusion due to a concentration gradient, for which a similar analysis can be performed). On the other hand, at each time instant, t , and for each point x , the law of energy conservation (neglecting heat generation) lauds (for other fields one must use the corresponding continuity equation)

$$-divq(x, t) = \rho c \frac{\partial T(x, t)}{\partial t} \quad (7)$$

where ρ is the density and c is the specific heat.

From Eqs. (6) and (7) we can obtain the so-called hyperbolic heat equation

$$\frac{\partial^2 T(x, t)}{\partial x^2} - \frac{1}{\alpha} \frac{\partial T(x, t)}{\partial t} - \frac{1}{u^2} \frac{\partial^2 T(x, t)}{\partial t^2} = 0 \quad (8)$$

CASE I: $u \gg D \beta$

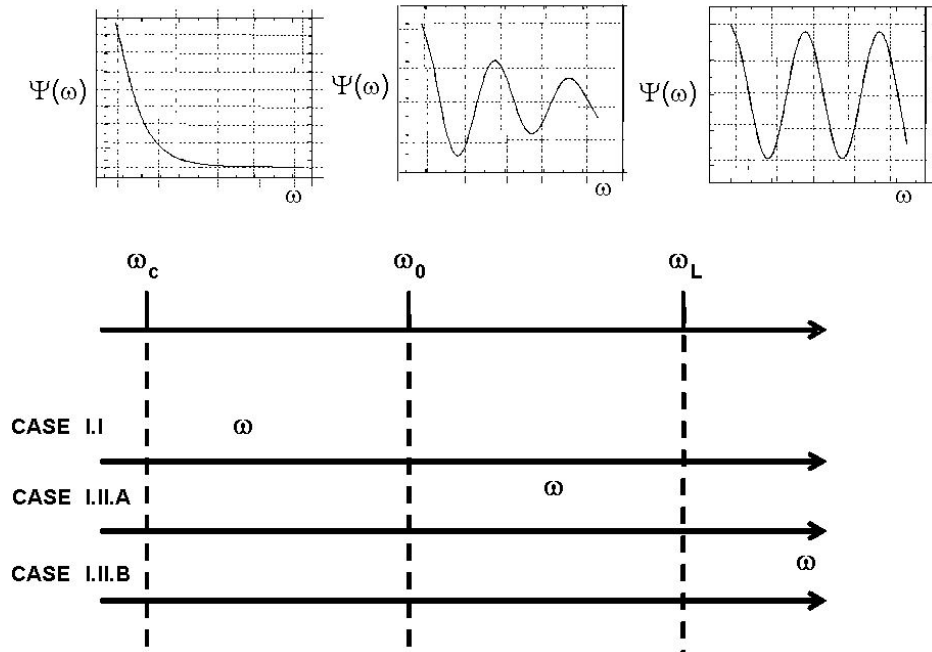


FIGURE 1. Schematic representation of the particular case I. At the top the form of the solutions of the generalized heat diffusion equation with a periodic source is shown schematically. For lower frequencies we have non-wave behavior. For frequencies between ω_0 and ω_L damped harmonic waves appears while for higher frequencies the waves are non-attenuated, and they propagate through a ‘non-dissipating’ medium (Although, not medium exists with zero resistance, an equivalent situation can be obtained in practice by choosing the adequate modulation frequency [10]).

CASE II: $u \ll D \beta$

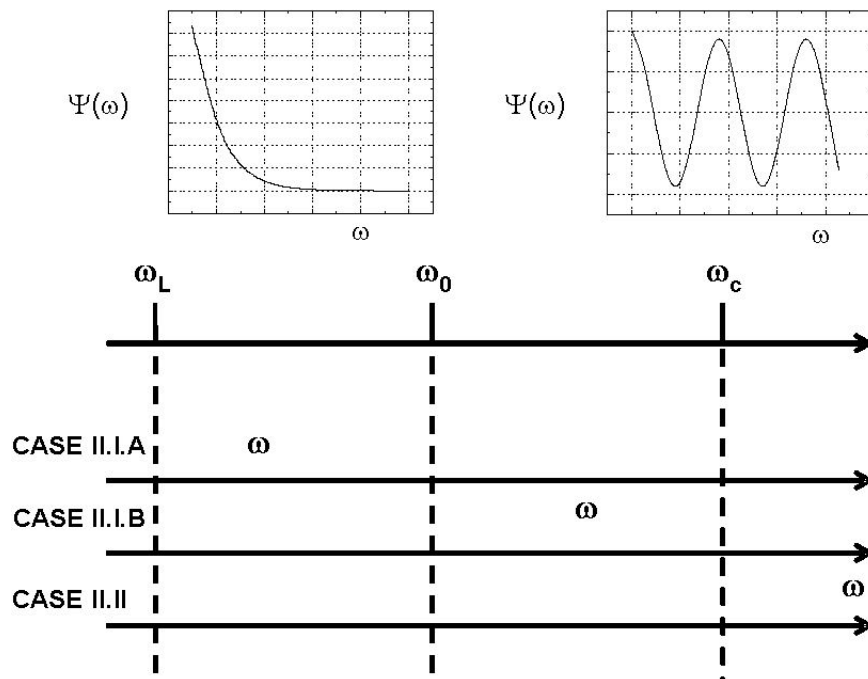


FIGURE 2. Schematic representation of the particular case II. At the top the form of the solutions of the generalized heat diffusion equation with a periodic source is shown schematically. For lower frequencies we have non-wave behavior. For frequencies above ω_0 non-attenuated harmonic waves appears.

where $\alpha = k/\rho c$ is the thermal diffusivity. The last term on the left hand side of this equation may help to solve the paradox of instantaneous heat propagation, introducing the velocity u , related to the relaxation time and to the thermal diffusivity through the relationship, $u = (\alpha / \tau)^{1/2}$, whose physical mean in the context of thermal waves has been discussed in detail elsewhere [10,13]. In some works a phase lag in the temperature gradient vector was also introduced to allow either the temperature gradient to precede the heat flux and vice-versa, introducing a term proportional to the time derivative of the second spatial derivative of the temperature field [15-17]. This is the so-called dual-phase-lag-model. A comparison between the results of the parabolic diffusion model, the hyperbolic one and the dual phase lag model has been performed elsewhere [18-20] for different cases of interest. As expected, the differences between the three models increase with the modulation frequency, due to of the effect of the time delays between heat flux and temperature gradient. It is a well-established fact that the dual-phase model becomes a necessity for very high frequencies of the heating source (for example for metal slabs they are larger than 10^{12} s^{-1}) [20]. Here, we shall limit our analysis to the classical hyperbolic case considering only the thermal relaxation time, τ .

We can see that Eq. (3) represents a generalized equation that can be used for the description of any kind of time varying diffusion phenomena. For large time scales compared with the relaxation times, the second derivative term can be neglected. This is the situation that most often appears in praxis in phenomena such as the above-described (mass, carrier and temperature diffusive fields). Care must be taken, however, in dealing with very fast processes such as those that would be induced by ultra-short duration laser pulses that can lead to situations where such physical-mathematical formalism is no longer valid and where the second time derivative must be taken into account.

In the presence of a periodic source of the form $Q(x)e^{i\omega t}$, the solution of physical interest of the problem for the applications considered here is that related to the time-dependent component of the solution to Eq. (3). If we separate this component from the spatial distribution, the field magnitude can be expressed as:

$$\varphi(x, t) = \text{Re} [\Theta(x) \exp(i\omega t)] \quad (9)$$

Substituting in Eq. (3), we obtain, for the spatial component, the equation:

$$\frac{d^2\Theta(x)}{dx^2} - q^2\Theta(x) = 0, \quad (10)$$

where

$$\begin{aligned} q^2 &= \left(\frac{\omega}{u}\right)^2 \left\{ \frac{\omega_C \omega_L}{\omega^2} \left[1 + i \frac{\omega}{\omega_C} \right] - 1 \right\} \\ &= \left(\frac{\omega}{u}\right)^2 \left\{ \left(\frac{\omega_0}{\omega}\right)^2 \left[1 + i \frac{\omega}{\omega_C} \right] - 1 \right\} \end{aligned} \quad (11)$$

is the square of the complex diffusion wave number, q , depending on the characteristic frequencies:

$$\omega_C = D\beta^2 \quad (12)$$

$$\omega_L = \frac{u^2}{D} \quad (13)$$

$$\omega_0 = \sqrt{\omega_L \omega_C} = u\beta \quad (14)$$

Equation (11) represents the dispersion relation of the problem being considered. The solution of Eq. (10) depends, therefore, on the relative value of the modulation frequency, ω , respecting the characteristic frequencies given by Eqs. (12) to (14). Different particular cases will be analyzed below.

3. Particular cases and discussion

We shall discuss the possible particular cases, which are illustrated in Figs. 1 and 2.

Case I: $\omega_c < \omega_0 < \omega_L$

This occurs for $u \gg D\beta$ as we can easily see from Eqs. (12) to (14) (Fig. 1). This is a common practical situation, as we shall discuss later in this section.

Case I.I: $\omega \ll \omega_C$

One has

$$q^2 = \left(\frac{\omega}{u}\right)^2 \left\{ \left(\frac{\omega_0}{\omega}\right)^2 - 1 \right\} \quad (15)$$

As $\omega \ll \omega_0$

$$q^2 = \beta^2 \quad (16)$$

The solution to Eq. (10) has the form

$$\Theta_{IA}(x) = \Theta_{IA0} \exp(-\beta x), \quad (17)$$

i.e., there are no waves for frequencies ω such that $\omega \ll \beta u$. In the above expression, as well as in the following equations, the explicit form of the amplitude term (here Θ_{IA0}) is not relevant for the purposes of this work (the frequency dependent amplitude terms can be obtained using proper boundary conditions. Generally the Θ_0 s decrease as ω increases).

Case I.II: $\omega \gg \omega_c$

One becomes

$$q^2 = \left(\frac{\omega}{u}\right)^2 \left[i \frac{\omega_L}{\omega} - 1 \right] \quad (18)$$

Case I.II.A: $\omega \ll \omega_L (\omega_L \gg \omega_C)$

The wave number becomes

$$q = \sqrt{i \frac{\omega}{D}} = \frac{(1+i)}{\mu} \quad (19)$$

i.e. their real and imaginary parts are equal to the inverse of the frequency dependent length $\mu=(2D/\omega)^{1/2}$. The solution to Eq. (10) is an attenuated wave given by:

$$\Theta_{IIA}(x) = \Theta_{IIA0}(\omega) \exp\left(-\sqrt{\frac{\omega}{2D}}x\right) \times \exp\left(-i\sqrt{\frac{\omega}{2D}}x\right) \quad (20)$$

This practical and very important case represents a mode through which energy is transferred to the surrounding media by diffusion at a rate determined by diffusion coefficient D . The characteristic (diffusion) length, μ , gives the distance at which the propagated wave amplitude decays e times from its value at the surface. It can be interpreted as a kind of skin depth depending on the diffusion coefficient, D , the relevant parameter for time-dependent diffusion processes within homogeneous, isotropic materials. (It is similar to the skin depth in a metal, $\mu_e=(2/\omega_p p \sigma)^{1/2}$, for electromagnetic waves in an electrically conductive medium. In that case, as is well known, a dispersion relation is defined assigning the photons a frequency, ω_p , depending on the wave number k_e as

$$k_e = \left((\omega_p/c)^2 + i\omega_p p \sigma \right)^{1/2},$$

where c is the velocity of light in the medium with permeability p and electrical conductivity σ). The diffusion length can be varied experimentally by changing the modulation frequency, ω , permitting depth profiling. Equation (20), therefore, represents an attenuated wave. Between the excitation and the response of the sample there is a phase-lag given by the term (x/μ) in the complex exponent.

Case I.II.B: $\omega \gg \omega_L (\omega_L > \omega_C)$

Now the wave number becomes $q=i\omega/u$, and the solution to Eq. (10) is a non-attenuated, harmonic wave:

$$\Theta_{IIB}(x) = \Theta_{IIB0}(\omega) \exp\left(-i\frac{\omega}{u}x\right) \quad (21)$$

At a given frequency ω , it travels across any sample without attenuation and with velocity u .

By analyzing the results obtained for these cases, we can reach the following conclusions. Until a critical frequency $\omega_C = D\beta^2$ is reached, there are no wave fields propagating through the sample. The energy transfer is governing by a classical diffusion equation. The frequency ω_C can be interpreted as the inverse of a characteristic time after which the energy carrier “disappears” in an energy conversion process. This time can be for example the carrier recombination life-time in a semiconductor, the time after which a propagating photon is absorbed in a turbid medium or the time after which a heat carried phonon gives its energy to another

particle interacting with it. Once this frequency is achieved in the modulation process, the medium becomes resistive, an attenuated wave field is imposed, and the characteristic decay length μ decreases with the increase in modulation frequency. When the frequency equals the value $\omega_L = u^2/D$, the material becomes transparent to the propagating wave field, and a non-attenuated wave propagates through the sample with the velocity u . This situation is equivalent to the hypothetical case of a non-resistive medium, *i.e.* one with an infinite diffusion coefficient, $D \rightarrow \infty$, where the transport of energy loses its diffusive character (the equivalent situation for electromagnetic waves is the case of a non-dispersive medium, *i.e.* a medium with negligible electrical conductivity, such as a vacuum). This wave can travel across the sample without attenuation and with velocity u .

As mentioned above, the situation where $u \gg \beta$ is often encountered in praxis. For example, for (hyperbolic) thermal waves, u is of the order of the sound velocity in the medium [10], and $\beta = 0$. In this particular case, we have neither ω_0 nor ω_C . This is the case of the thermal wave field discussed in detail elsewhere [10]. It appears as the solution to the heat diffusion equation in the presence of modulated heat sources. For the frequencies often encountered in PA and PT experiments, we have attenuated parabolic waves. A typical example which illustrates this particular case is the so-called thermal wave. Although nowadays the thermal wave concept has become of great interest for the explanation and interpretation of photothermal and photoacoustic phenomena and techniques [21], it is worth mentioning here that several authors have demonstrated that there is no wave nature in this concept by showing that parabolic thermal waves do not transport energy [22] and by demonstrating the reflection- and refractionless nature of parabolic thermal wave fields [23]. However, as mentioned above, the analogy with waves enables the description of a number of phenomena related to time varying heat transfer phenomena, some of which are described in detail elsewhere [24]. In experiments performed under (now idealized) conditions such that frequencies $\omega \gg \omega_L$ can be obtained, one can become a hyperbolic non-attenuated wave field in which phenomena such as second sound propagation in solids can be studied [10,13-14,25,26]. Hyperbolic heat transport has also received increasing attention for the analysis of practical problems involving a fast supply of thermal energy (for instance heating of materials with intense ultra-short-duration laser pulses [27], transient hot wire measurements of thermal conductivity in nanofluids [28], gravitational collapse of stars [29], and many others), as well as due to theoretical motivations regarding the peculiarities of hyperbolic heat propagation in different media and under dissimilar conditions [30-33]. In this case, the characteristic time constant can also be related to the “memory” effects of the corresponding flux for systems where the propagation speeds are small (economic and biological systems have been described elsewhere [34] using diffusion equations, and they are examples

where memory effects are quite relevant, since humans make decisions according to their previous experiences).

On the other hand, when we set $\beta \neq 0$ and $\partial^2/\partial t^2 \rightarrow 0$, it becomes the typical situation encountered in plasma wave experiments with semiconductors, photon diffusion through dispersive media, etc, where $\hat{F}_n = \beta^2$, *i.e.* the square of the inverse of a typical length, as described in Ref. 1. In this case $\omega_L \rightarrow \infty$, and we have an attenuated wave whenever $\omega > \omega_C$.

Case II.: $\omega_L < \omega_0 < \omega_C$

It is illustrated in Fig. 2 and takes place for $u \ll D\beta$. Although it is a less common situation, the analysis of this particular case can also be of interest.

Case II.I: $\omega \ll \omega_C$

One has, as in case I.I

$$q^2 = \left(\frac{\omega}{u}\right)^2 \left\{ \left(\frac{\omega_0}{\omega}\right)^2 - 1 \right\} \quad (22)$$

Case II.I.A: $\omega \ll \omega_0$

$$q^2 = \beta^2 \quad (23)$$

The solution to Eq. (10) has the form given by Eq. (17).

Case II.I.B: $\omega \gg \omega_0$

Now the wave number becomes $q=i\omega/u$, and the solution to Eq. (10) is a non-attenuated, harmonic wave, which is described by Eq. (21).

Case II.II: $\omega \gg \omega_c$ ($\omega \gg \omega_L, \omega \gg \omega_0$)

As in case II.I.B, the wave number becomes $q=i\omega/u$, and the solution to Eq. (10) has the form given by Eq. (21).

We have seen that for $u \ll D\beta$, one has only a (non-attenuated, harmonic) wave field whenever we exceed the critical frequency ω_c . This field propagates through a transparent material with velocity u . For lower frequencies, there are no waves. The field magnitude is independent of the modulation frequency and undergoes attenuation determined by the value of the β parameter [see Eq. (17)]. Note that for frequencies much lower than ω_0 , the wave number becomes zero [see Eq. (11)] and the Diffusion Equation [Eq. (10)] becomes the Laplace Equation, with a linear decreasing dependence of the field parameter on the coordinate x . There is no frequency dependence in the field magnitude either.

In summary: for both particular cases analyzed we have no waves for frequencies lower than $\omega_0 = u\beta$ and we have harmonic waves for higher frequencies. These are always non-attenuated waves propagating at velocity u for the case in which $u \ll D\beta$. For the much more typical situation

where $u \gg D\beta$, there are attenuated thermal waves for frequencies lower than $\omega_L = u^2/D$, at which the material becomes transparent, permitting wave propagation without attenuation. These behaviors are shown qualitatively in the top parts of Figs. 1 and 2. Note that the amplitude of the signal always decreases as ω increases.

4. Conclusions

In summary, the phenomenological aspects described here suggest the possibility of dealing in advanced or introductory physics courses with concepts related to diffusion waves. Although numerous attempts have been made to give a phenomenological explanation of different kinds of diffusion waves, a generalized equation analyzed here should be of great pedagogical interest. Since this equation describes many kinds of diffusion phenomena, it can be applied to all of them, with the proper interpretation of each particular physical situation. Although extensive theoretical work has been published before in the field of diffusion wave fields (the majority of them related to parabolic and hyperbolic heat diffusion), and several attempts to give analytical solutions of the involved equations exist for different cases of practical and academic interest, the discussion of the limiting cases of the corresponding dispersion equation in the frequency domain performed here, and the qualitative, phenomenological discussion of the solutions to the wave equation in the case of periodical sources, should be significant for teaching purposes. The discussion of the analytical solutions to the proposed equation must be limited to special cases and it is beyond the scope of this article. Thus we feel that the approach presented in this study is plausible and satisfying without involving excessively sophisticated mathematical techniques. Although part of the questions discussed here is not new, hopefully the basic ideas presented here will aid in opening the literature associated with this theme to a wider audience. This work represents a step towards the consolidation of this objective and towards the better interpretation of periodically excited diffusion phenomena. It is worth remembering, before concluding, that the first wave treatment of periodical phenomena dates from the 1820's when Fourier [35] showed that heat conduction problems could be solved by expanding temperature distributions as series of waves. Two centuries later, the concept of diffusion waves still receives the attention of scientists from several fields of research.

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