Investigating Electromagnetic Pulses in Matter

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1 Introduction

Certain electromagnetic pulses are capable of altering materials in amazing ways as they propogate through them, possibly even breaking particular chemical bonds by vibrating them but not others. As part of ongoing research whose final and highest aim is to accomplish feats like this, we have set up a control experiment in which controllable light pulses propogate through a glass cylinder containing a particular solution. The goal of our experiment is to shape the pulse such that the wave intensity takes some interesting form (i.e. intensity is even throughout, higher after travelling through absorbtive medium, etc). The tube is probed with weak electromagnetic in a few places along its length, and each probe has a sensor that reads the probe after it has interacted with the matter. These sensors are connected to a computer with a genetic algorithm that determines, based on a series of past experiments, the shape of the light pulse for the next trial.

I will be concentrating on the electromagnetic aspects of the experiment. This is obviously just one aspect of the experiment, as it also involves quantum mechanics, chemistry, and computer science. Though much work has been done that relates to electromagnetic wave propogation in matter, much less has been done from a control perspective. For this reason, it is necessary to define and answer the following questions.

1.1 Questions to be addressed:

Taken from Alex's notes:

1. Determine what is the physically admissible dependence of permittivity $\epsilon_{i;j}(\omega)$, $\epsilon_{i;kl}(\omega_1, \omega_2)$ on frequencies ω_1, ω_2...? Classify functional forms and magnitudes of permittivity for each type of medium. For example, which type of medium corresponds to non-zero $\epsilon_{i;kl}(\omega_1, \omega_2)$? When can the permittivity be approximated by a constant, i.e. when can one can neglect dispersion? When can we neglect memory effects in the field in the expansion (2)? When is dependence of permittivity on indices i; j; k; l non-trivial?

2. Set up the mathematical problem which corresponds to the picture above. This includes:

2a. Derive the wave equation for electric field E propagating in a medium with given permittivity. This can be obtained by substitution of the expansion (2) into equation (1). The wave equation in general will contain a nonlinear term. Justify the physical validity of approximation grad div E = 0. Are there other "reasonable" approximations?

2b. Set up the boundary conditions for the picture above.

2c. Set up the relation between the material property and the mapping $S_i \rightarrow S_f$

i. This mapping will go into forming the cost function.

2d. Define the cost function.

3. Determine an appropriate method for solving numerically the control problem for fixed permittivity. It can be either a GA or a gradient algorithm.

4. Solve the control problem.

5. Determine how the controllability depends on permittivity. Find general properties of the permittivity which are helpful/destructive for the control.

2 Maxwell's Equations in Matter

2.1 Polarization and Magnetization

We know Maxwell's equations in a vacuum:

$$(1) \qquad \nabla \cdot E = 4\pi\rho \qquad\qquad \nabla \cdot B = 0$$

$$(2) \qquad \nabla \times E = -\frac{1}{c}\frac{\partial B}{\partial t} \qquad\qquad \nabla \times B = \frac{4\pi}{c}J + \frac{1}{c}\frac{\partial E}{\partial t}$$

In matter, we have to consider that matter is polarized and magnetized by electromagnetic radiation. Consider the following two facts:

a) a changing P results in a current density $J_p = \frac{\partial P}{\partial t}$

b) a circulation of M results in a current density $J_m = c\nabla \times M$

To understand how J_p comes about, consider group of N molecules, each with dipole moment p. p is a vector with units of *charge* $\times$ *distance*. Then the macroscopic polarization is the quantity P=Np. Now, imagine that during a period of time dt, the dipole moment of each molecule changes by dp (which could result from either a reorganization of the charge in the molecules of a shift in their position). The change dp could have been caused by moving a charge q a distance d, and we would have $dp = q\,dd$ (the change could have resulted from another cause, but the situation is analagous and this description is perfectly valid).

We can see that a conduction current J_p arises because

$$(3) \qquad J_p = Nq\frac{dd}{dt} = N\frac{dp}{dt} = \frac{dP}{dt}$$

Though the current arises from stuctural reorganizations of the molecules, it is nonetheless moving charge, and is thus the equivalent of moving free charges.

Figure 1: curl M gives J

We wondered why the polarization would matter, given that the field was changing so quickly compared to the response time of the molecules. As Professor Rabitz pointed out, the electrons respond instantaneously to the field, but the molecules' response time can be reduced by friction between molecules. In nonpolar molecules, the response is very fast because very little mass is involved in the reorganization of electrical structures. In polar media, however, the whole molecule must rotate, making the response time much slower. In water, for example, the dialectric constant ϵ is around 80 (meaning P is quite significant) unless the field oscillates with frequencies around and above 10^{11} Hz or higher, in which case ϵ drops to about 5. This is due to the fact that the molecules simply cannot reorient themselves quickly enough to contribute much polarization when the field changes in 10^{-11} seconds or less.

To see how the magnetization M arises, consider a material with a magnetization which is not constant in space. WLOG, assume that the magnetization increases along the y axis, as pictured.

Now there is a total current density described by $J_{free} + \frac{\partial P}{\partial t} + J_m = c\nabla \times M$. Further, polarization introduces a charge density ρ_{bound} so Maxwell's equations would read:

$$(4) \qquad \nabla \cdot E = 4\pi(\rho_{bound} + \rho_{free}) \qquad\qquad \nabla \cdot B = 0$$

$$(5) \qquad \nabla \times E = -\frac{1}{c}\frac{\partial B}{\partial t} \qquad\qquad \nabla \times B = \frac{4\pi}{c}(J_{free} + c\nabla \times M + \frac{\partial P}{\partial t}) + \frac{1}{c}\frac{\partial E}{\partial t}$$

If we define the quantities H and D in their usual ways, where D (the *displacement current*) is E + 4πP and H (the so-called *auxiliary magnetic field*) is B - 4πM, we can write Maxwell's equations in terms of ρ_{free} only:

$$(6) \qquad\qquad \nabla \cdot D = 4\pi\rho_{free} \qquad\qquad \nabla \cdot B = 0$$

$$(7) \qquad\qquad \nabla \times E = -\frac{1}{c}\frac{\partial B}{\partial t} \qquad\qquad \nabla \times H = \frac{4\pi}{c}J_{free} + \frac{1}{c}\frac{\partial D}{\partial t}$$

2.2 Memory Effects

These equations account for polarization and magnetization, but there is more to the story when fields that change in time are introduced. Changes in magnetization and polarization do not happen instantaneously when acted upon by an electric field. Rather, the field causes accelerations, not instantaneous adjustment, and for this reason magnetization and polarization depend on the history of electromagnetic field presense in the matter. The simple proportionalities

$$H \propto B \qquad P \propto E$$

are not valid when memory effects are considered. If we consider the inertia of the electrons in matter, we have a space and time dependent polarization given by

$$(8) \qquad\qquad \vec{P}(\vec{r},t) = \int_{-\infty}^{t} \chi(\vec{r}, t - t')E(\vec{r}, t')dt'$$

To do a Fourier transform of this function, we define the susceptibility $\chi(\vec{r}, t)$ to be 0 for $t < 0$, making the above relation read

$$(9) \qquad\qquad \vec{P}(\vec{r},t) = \int_{-\infty}^{\infty} \chi(\vec{r}, t - t')E(\vec{r}, t')dt'$$

We must also let

$$(10) \qquad \widetilde{\chi}(\vec{r},\omega) = \int_{-\infty}^{\infty} \chi(\vec{r}, t)e^{i\omega t}dt \qquad\qquad \widetilde{E}(\vec{r},\omega) = \int_{-\infty}^{\infty} E(\vec{r}, t)e^{i\omega t}dt$$

Now we can express P in terms of a frequency ω as

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$$(11) \qquad \tilde{P}(\vec{r}, \omega) = \int_{-\infty}^{\infty} P(\vec{r}, t) e^{i\omega t} dt$$

$$(12) \qquad = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{i\omega t} \chi(\vec{r}, t - t') E(\vec{r}, t') dt' dt$$

$$(13) \qquad = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{i\omega(t-t')} \chi(\vec{r}, t - t') e^{i\omega t'} E(\vec{r}, t') dt' dt$$

$$(14) \qquad = \int_{-\infty}^{\infty} e^{i\omega t''} \chi(\vec{r}, t'') dt'' \int_{-\infty}^{\infty} e^{i\omega t'} E(\vec{r}, t') dt'$$

$$(15) \qquad = \tilde{\chi}(\vec{r}, \omega) \tilde{E}(\vec{r}, \omega)$$

We see that there is still a linear relationship between P and E for each frequency. Since the relationships between P and E and between B and M (and thus M and H) are analogous, we can also say that $\widetilde{M}(\vec{r}, \omega) = \widetilde{\chi_m}(\vec{r}, \omega) \tilde{H}(\vec{r}, \omega)$

Maxwell's equations, for frequency dependent $\widetilde{\chi_e}$ and $\widetilde{\chi_m}$, are

$$(16) \qquad \nabla \cdot \tilde{D}(\vec{r}, \omega) = 4\pi \rho_{free} \qquad\qquad \nabla \cdot \tilde{B}(\vec{r}, \omega) = 0$$

$$(17) \qquad \nabla \times \tilde{E}(\vec{r}, \omega) = \frac{i\omega}{c} \tilde{B}(\vec{r}, \omega) \qquad\qquad \nabla \times \tilde{H}(\vec{r}, \omega) = \frac{4\pi}{c} J_{free} - \frac{i\omega}{c} \tilde{D}(\vec{r}, \omega)$$

Where $\tilde{H}(\vec{r}, \omega) = \tilde{B}(\vec{r}, \omega) - 4\pi \widetilde{M}(\vec{r}, \omega)$ and $D(\vec{r}, \omega) = \epsilon(\vec{r}, \omega) E(\vec{r}, \omega)$.

2.3 The Wave Equation

A straightforward arranging of equations 16 and 17 prescribes the following equation:

$$(18) \qquad \nabla \times (\nabla \times \tilde{E}) = \nabla(\nabla \cdot \tilde{E}) - \nabla^2 \tilde{E} \qquad\qquad \text{vector identity}$$

$$(19) \qquad = \nabla \times (\frac{i\omega}{c} \tilde{B}(\vec{r}, \omega)) \qquad\qquad \text{substitute } \frac{i\omega}{c} \tilde{B}(\vec{r}, \omega) \text{ for } \nabla \times \tilde{E}$$

$$(20) \qquad = \frac{i\mu\omega}{c} (\nabla \times \tilde{H}) \qquad\qquad \text{assume } \mu \text{ is constant s.t. } \tilde{B}(\vec{r}, \omega) = \mu \tilde{H}(\vec{r}, \omega)$$

$$(21) \qquad = -\frac{\omega^2 \mu}{c} \tilde{D}(\vec{r}, \omega) \qquad\qquad \text{substitute for } \nabla \times \tilde{H}(\vec{r}, \omega)$$

$$(22)$$

which is the wave equation

$$(23) \qquad \nabla^2 \tilde{E} = \frac{\omega^2 \mu}{c} \tilde{D}(\vec{r}, \omega)$$

if μ is assumed to be constant and we can assume that $\nabla(\nabla \cdot E) \approx 0$. We know that this is true because $\nabla \cdot E$ is the intensity of the wave, and the change in the intensity of the wave over a small distance is small compared to $\nabla^2 E$, or the second spacial derivitive of the E field. This approximation is called the "slowly varying envelope" approximation.

In a continuous anisotropic media, D_i can be expressed as a sum of powers of E as

$$(24) \qquad \widetilde{D} = \widetilde{D}_i^{(1)}(\omega) + \widetilde{D}_i^{(2)}(\omega) + \dots$$

where

$$(25) \qquad \widetilde{D}_i^{(1)}(\omega) = \sum_{j=1}^{3} \chi_{i;j}(\omega) \widetilde{E}_j(\vec{r}, \omega)$$

and

$$(26) \qquad \widetilde{D}_i^{(2)}(\omega) = \sum_{k,l=1}^{3} \int \chi_{i;kl}(\omega - \omega', \omega) \widetilde{E}_k(\vec{r}, \omega - \omega') \widetilde{E}_l(\vec{r}, \omega') d\omega'$$

In our case, we have EM waves which are polarized in the same axis, i.e.

$$(27) \qquad E^c(x, t) = (0, 0, E_z^c(x, t))$$

and

$$(28) \qquad E^p(y, t) = (0, 0, E_z^p(y, t))$$

Thus, we define $\chi_{i;j}$ as

$$\chi_{i;j} = \begin{cases} \chi_0(\omega) & \text{if } i = j \\ 0 & \text{else} \end{cases}$$

and $\chi_{i;kl}$ as

$$\chi_{i;kl} = \begin{cases} \chi_1(\omega, \omega - \omega') & \text{if } i = k = l \\ 0 & \text{else} \end{cases}$$

Thus, there is one non-zero $\chi(\omega)$ in the linear term and one non-zero $\chi(\omega, \omega - \omega')$ in the first nonlinear term in the expansion of D, so the expansion of $D_{total} = D_{control} + D_{probe}$ will have the basic form:

$$(29) \qquad D_{total} \approx D_{total}^{(1)} + D_{total}^{(2)}$$
$$(30) \qquad = E_{total} + \chi(\omega) E_{total} + \chi(\omega, \omega - \omega') E_{total}^2 +$$
$$(31) \qquad = (E_c + E_p) + \chi(\omega)(E_c + E_p) + \chi(\omega, \omega - \omega')(E_c + E_p)^2$$
$$(32) \qquad = (E_c + E_p) + \chi(\omega)(E_c + E_p) + \chi(\omega, \omega - \omega')(E_c^2 + E_p^2 + 2E_c E_p)$$

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Here we can see that the reason that there is any interaction between the control pulse and the probe pulse, and hence the reason that the experiment works at all, is the nonlinear term in the expansion for D. If we were just to consider the linear approximations to D for both pulses, then the effects that the two pulses have on one another would not be included in the EM equations at all. The memory effects must be considered for they are the source of the field's interaction.

The control field is much stronger than the probe field, so in formulating its wave equation, we can ignore the effects of the probe field (but not the first nonlinear term of E_c). Because the fields of E_c and E_p are superpositioned in equations 30-32, we can separate the terms of E_c and E_p when writing their respective wave equations. From equation 32 we can find the approximate wave equation for $E_c(\vec{x}, \omega)$:

$$\frac{\partial^2 E_c(\vec{x}, \omega)}{\partial x^2} = \frac{\omega^2 \mu}{c} \left[E_c(\vec{x}, \omega) + \chi_0(\omega) E_c(\vec{x}, \omega) + \right.$$
$$\left. + \int d\omega' \, \chi_1(\omega - \omega', \omega) E_c(\vec{x}, \omega - \omega', \omega) E_c(\vec{x}, \omega') + \dots \right]$$
(33)

and for E_p:

$$\frac{\partial^2 E_p(\vec{x}, \vec{y}, \omega)}{\partial y^2} = \frac{\omega^2 \mu}{c} \left[E_p(\vec{y}, \omega) + \chi_0(\omega) E_p(\vec{y}, \omega) + \right.$$
$$+ \int d\omega' \, \chi_1(\omega - \omega', \omega) \left\{ E_p(\vec{y}, \omega - \omega', \omega) E_p(\vec{y}, \omega') + \right.$$
$$\left. \left. + E_c(\vec{x}, \omega - \omega') E_p(\vec{y}, \omega') + E_p(\vec{y}, \omega - \omega') E_c(\vec{x}, \omega') \right\} + \dots \right]$$
(34)

The terms in this expansion which have both E_p and E_c can be combined by the introduction of a new variable of integration ω'' where $\omega'' = \omega - \omega'$. Observe that the last line in equation 34:

$$\text{(35)} \qquad \int d\omega' \, \chi_1(\omega - \omega', \omega) \left\{ E_c(\vec{x}, \omega - \omega') E_p(\vec{y}, \omega') + E_p(\vec{y}, \omega - \omega') E_c(\vec{x}, \omega') \right\}$$

can be written

$$\text{(36)} \quad \int d\omega' \, \chi_1(\omega - \omega', \omega) E_c(\vec{x}, \omega - \omega') E_p(\vec{y}, \omega') - \int d\omega'' \, \chi_1(\omega'', \omega) E_c(\vec{x}, \omega - \omega'') E_p(\vec{y}, \omega'')$$

giving the control term

$$\text{(37)} \qquad \widetilde{\chi}(\vec{x}, \omega, \omega') = \Big(\chi_1(\omega - \omega', \omega) - \chi_1(\omega', \omega) \Big) E_c(\vec{x}, \omega - \omega')$$

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such that

$$\frac{\partial^2 E_p(\vec{x}, \vec{y}, \omega)}{\partial y^2} = \frac{\omega^2 \mu}{c} \Big[E_p(\vec{y}, \omega) + \chi_0(\omega) E_p(\vec{y}, \omega) +$$

$$+ \int d\omega' \, \chi_1(\omega - \omega', \omega) E_p(\vec{y}, \omega - \omega', \omega) E_p(\vec{y}, \omega') +$$

(38)
$$+ \int d\omega' \, \widetilde{\chi}(\vec{x}, \omega, \omega') E_p(\vec{y}, \omega') + \dots \Big]$$

Now we need boundary conditions for our specific case.

2.4 Boundary Conditions

To simplify things in order to formulate a working equation, we begin by making the assumption that the E_c^2 term is negligable in the wave equation for E_c. We can solve the wave equation because we know the initial conditions and control the boundary condition $E_c(x_0, t)$. Here, x_0 is the (x) position at which the pulse enters the media, and we can consider the experiment as if it were done in a semi-infinite tube with with origin x_0.

For the waveform $E(x, t)$ propogating in the positive x direction with speed v and with initial and boundary conditions

$$E(x, 0) = 0 \qquad \frac{\partial E}{\partial t}(x, 0) = 0 \qquad E(0, t) = \xi(t)$$

the solution is straightforward:

(39)
$$E(x, t) = \xi(t - \frac{x}{v})$$

The value of $E(x, t)$ depends only on the boundary function's value at the time when the wave-value originated. $E(x, t)$ will be of the form $f(x - vt)$ because the waveform will only be propogating to the right. In cases where dispersion is present, we can have many $\xi_i(0, t)$ for each frequency, each having its own corresponding speed of propogation, and we can say:

(40)
$$E(x, t) = \sum_{v_i} \xi_i(t - \frac{x}{v_i})$$

Each ξ_i will have the form of a sinusoidal function, which can be represented by $e^{i\omega(t - \frac{x}{v_i})}$. Since $v = \frac{1}{\epsilon(\omega)}$, we have

(41)
$$E(x, t) = \sum_{v_i} A_i e^{i\omega(t - \sqrt{\epsilon(\omega_i)}x)}$$

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In integral form, this is

$$(42) \qquad E(x,t) = \int d\omega A_w e^{\imath\omega(t-\sqrt{\epsilon(\omega)}x)}$$

3 Summary and Working Equations

The wave equations for E_c and E_p:

$$(43) \qquad \frac{\partial^2 E_c(\vec{x},\omega)}{\partial x^2} = \frac{\omega^2\mu}{c}\Big[E_c(\vec{x},\omega) + \chi_0(\omega)E_c(\vec{x},\omega)\Big]$$

$$(44) \qquad = \frac{\omega^2\mu}{c}\epsilon(\omega)E_c(\vec{x},\omega) \text{ where } \epsilon(\omega) = 1 + \chi_0(\omega)$$

$$(45) \qquad \frac{\partial^2 E_p(\vec{x},\vec{y},\omega)}{\partial y^2} = \frac{\omega^2\mu}{c}\Big[E_p(\vec{y},\omega) + \chi_0(\omega)E_p(\vec{y},\omega) + \int d\omega'\ \tilde{\chi}(\vec{x},\omega,\omega')E_p(\vec{y},\omega')\Big]$$

$$(46) \qquad = \frac{\omega^2\mu}{c}\Big[\epsilon(\omega)E_p(\vec{x},\omega) + \int d\omega'\ \tilde{\chi}(\vec{x},\omega,\omega')E_p(\vec{y},\omega')\Big]$$

where

$$(47) \qquad \boxed{\tilde{\chi}(\vec{x},\omega,\omega') = \Big(\chi_1(\omega-\omega',\omega) - \chi_1(\omega',\omega)\Big)E_c(\vec{x},\omega-\omega')}$$

In general,

$$(48) \qquad \boxed{E(x,t) = \int d\omega A_\omega e^{\imath\omega(\sqrt{\epsilon(\omega)}x - t)}}$$

Consider the boundary condition that $E(0,t) = E_0(t) = \int d\omega A_\omega e^{-\imath\omega t}$, we have

$$(49) \qquad \boxed{A_\omega = \frac{1}{2\pi}\int dt E_0(t)e^{\imath\omega t}}$$

The initial and boundary conditions for E_c are

$$E_c(x,0) = 0 \qquad \frac{\partial E_c}{\partial t}(x,0) = 0 \qquad E_c(0,t) = \int d\omega A_\omega e^{-\imath\omega t}$$

The initial and boundary conditions for E_p are

$$E_p(y,0) = 0 \qquad \frac{\partial E_c}{\partial t}(y,0) = 0 \qquad E_p(0,x,t) = \int d\omega B_{x,\omega} e^{-\imath\omega t}$$

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4 Implementing a Control Algorithm

Now we are ready to consider the practicalities of implementing a useful computer algorithm using these equations. Begin by noting that the integrals in our waves equations will actually be sums of a number (somewhere in the neighborhood of 100) of terms.

To solve for the probe field using equation **??**, we can use an iterative process. First, assume that the $E_p E_c$ term is small and ignore it to solve for E_p^0. On the next iteration, we can use the value of E_p^0 in the $E_p E_c$ term and determine E_c from equation 48. We will then have E_p^1, and the process can be repeated in the same manner.

The form of equation **??** is that of a linear, nonhomogeneous differential equation with a constant term $\int d\omega' \; \widetilde{\chi}(\vec{x}, \omega, \omega') E_p(\vec{y}, \omega')$

Equations of this form can be solved analytically with a Green's function.