Real-time determination of free energy and losses in optical absorbing media

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We introduce notions of free energy and loss in linear, absorbing dielectric media which are relevant to the regime in which the macroscopic Maxwell equations are themselves relevant. As such we solve a problem eluded to by Landau and Lifshitz in 1958, and later considered explicitly by Barash and Ginsburg, and Oughtsun and Sherman. As such we provide physically-relevant real-time notions of "energy" and "loss" in all analogous linear dissipative systems.

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In a previous publication we showed that in many instances fast and slow light is a manifestation of a dielectric interacting differently with the early parts of an electromagnetic pulse than with its later parts. For example, slow light in a passive linear dielectric typically corresponds to energy in the leading part of the pulse being preferentially stored by the medium and then being largely returned to the pulse's (backward) tail. Quantifying the extent to which this process is possible was a primary motivation for this work.

Application of the principles presented in this letter will in no-wise supersede the group velocity description of such phenomena. The present development compliments other such analysis by attaching precise notions of reversibility and irreversibility to the medium's storage of the pulse energy. As such it also solves a long outstanding problem regarding the real-time meaning of "energy" and "loss" in linear dissipative systems.

From Poynting's energy conservation theorem (Eq. 1) the total energy density in a dielectric at time $t$, $u(t)$, is the sum (Eq. 2) of the field energy $u_{\text{field}}(t)$ (Eq. 3) and the medium-field interaction energy $u_{\text{int}}(t)$ (Eq. 4):

$$\nabla \cdot S + \frac{\partial u}{\partial t} = 0 \quad (1)$$

$$u(t) = u_{\text{field}}(t) + u_{\text{int}}(t) \quad (2)$$

$$u_{\text{field}}(t) = \frac{E^2}{2} + \frac{H^2}{2} \quad (3)$$

$$u_{\text{int}}(t) = \int_{-\infty}^{t} E(r) \dot{P}(r) \, dr. \quad (4)$$

Here we restrict to isotropic, temporally dispersive media in which a scalar analysis suffices, and in which one may safely suppress reference to the spatial coordinate $x$. In addition we restrict to linear media. Consequently we restrict to the case in which the medium's response to an applied field $E$ is completely determined by a scalar, point-wise defined susceptibility $\chi = \chi(\omega, x)$. For a basic discussion of properties of the medium-field interaction energy $u_{\text{int}}(t)$ in anisotropic media, see. Since $u_{\text{int}}(t)$ quantifies the net work the field has done against the polarized medium in the course of creating the current system state, in the following we will also call $u_{\text{int}}(t)$ the (medium) internal energy. In the sequel, notions of work done, and the ability to do work in the future, will become pivotal in establishing an unambiguous, dynamically relevant notion of energy allocation in dielectric media.

Landau and Lifshitz interpreted $u_{\text{int}}(+\infty)$ as the energy that is eventually lost to and dissipated by the medium over the course of the medium-field interaction. This asymptotic quantity only depends on the medium susceptibility $\chi$ and the evolution of the electric field $E$, not on the particulars of any microscopic model giving rise to $\chi$. This suggests the possibility of establishing a real-time, model independent notion of loss. Barash and Ginsburg considered this question. They concluded, however, that a real-time determination of losses is impossible without a microscopic model, i.e. that a knowledge of the macroscopic susceptibility $\chi$ is insufficient. Having concluded that a model independent notion of loss is meaningless, they calculated (a certain notion of) loss for specific models. In particular they generalized the work of Loudon concerning single Lorentz oscillator media by making a straightforward extension of his notion of losses to multiple Lorentz oscillators. Loudon’s and, so, Barash and Ginsburg’s notion of losses amounts to identifying those terms in (a certain expression of) the internal energy which explicitly depend on phenomenological damping parameters. More convincingly, their determination of the "energy" in the medium amounts to summing the kinetic and potential energies of the individual oscillators. We call this notion of energy the polarization energy, $u_{\text{polarization}}(t)$.

We initially followed a certain extension of this reasoning. We hoped to establish a unique map from an
arbitrary susceptibility $\chi(\omega)$ to an oscillator representation, which in turn would establish a unique (polarization) energy, as well as losses via Loudon’s identification procedure. Unfortunately, we found that generically a susceptibility can be mapped to many different microscopic models. This property is perfectly analogous to the one by which it is possible for distinct LRC circuits to yield identical impedances. However, we then conjectured that all such equivalent representations might yield identical values for the polarization energy. Unsurprisingly, one finds that this is not the case dynamically. (Although asymptotically all such representations must obviously agree: $u_{\text{polarization}}(+\infty) = 0$, and the losses are given, then, by $u_{\text{int}}(+\infty)$, which, as mentioned, is completely determined by $\chi$.)

Figures 1 and 2 demonstrate this ambiguity. Given a double Lorentz oscillator susceptibility $\chi(\omega) = \sum_{\nu=1,2} \omega_{\nu}^2 / (\omega_{\nu}^2 - i\gamma_{\nu}\omega - \omega^2)$ we examine two different microscopic models for the same susceptibility. The first model $\chi_a(\omega)$, is given by the explicit structure of $\chi(\omega)$ as just written. It corresponds to two different masses of equal charge on two different springs, each spring having different restoring and damping parameters. The total polarization is given by the net displacement of both charged masses from their equilibrium positions: $P = X_1 + X_2 = \chi_a E$. See FIG. 1(a).

We use the tangency algorithm [10], a transfer function preserving algorithm intended for model reduction in LRC circuits, to construct the second microscopic model.

\[ \chi_b(\omega) = \frac{1}{k_{1b} + \gamma_{1b} s + m_{1b} s^2 + \frac{1}{\gamma_{3b}} + \frac{1}{\gamma_{2b}/m_{3b} s^2}} \]  

By inserting the two representations into the internal energy $u_{\text{int}}(t) = \int_{-\infty}^{t} E(\tau) P(\tau) d\tau$ one can find the kinetic and potential energies associated with each representation. This polarization energy, the energy reckoned instantaneously via the motion and position of the masses, will be shown to be representation dependent. To demonstrate this, we calculate the “microscopic” energetics associated with the response of these macroscopically equivalent systems to an impulse. That is we excite the two media, with susceptibility models $\chi_a$ and $\chi_b$, via a delta function $E(\tau)$ at time $t = 0$, and then plot the losses for each representation. We assign values to the parameters of the two oscillator, $\chi_a$ representation (the plasma frequencies, resonant frequencies, and damping coefficients), and then find parameters for the coupled oscillators’, $\chi_b$ representation such that the representations are macroscopically equivalent. Figure 2 shows the evolution of the losses for the different representations, these losses determined in the sense of Loudon, Barash and Ginzburg. The corresponding differences between the internal energy imparted to the media by the delta function $E(\tau)$ (as shown by the piecewise constant curve in figure 2) and the plotted losses gives the polarization energies for the two representations.

The qualitative features of Figure 2 can be understood intuitively: the delta excitation of the medium instant-
taneously creates the polarization energies, which, then, decrease as the systems dissipate these energies. However, as clearly seen in the figures, the polarization energies for each microscopic representation differ significantly. Thus the polarization energy does in fact depend on the microscopic model ascribed to the susceptibility \( \chi(\omega) \). Of course, since Barash and Ginzburg specified no underlying macroscopic physical principle in their determination of "energy", this result is just a consequence of their (lack of a) definition of energy. Consequently, we will argue that a model-dependent (e.g. polarization) energy is irrelevant, both from the point of view of the relevant physical principles that should be required of a viable macroscopic description, as well as from that of the practical considerations of the energy storage and return process mentioned at the beginning of this article.

Before establishing relevant macroscopic principles of energy allocation, we highlight the connection between mechanical and electrical oscillators and the ambiguities associated with representation in the context of the latter. The perfect analogy between electrical LRC circuits and linear passive dielectric media (where polarization \( P \), susceptibility \( \chi \) and E-field \( E \), translate to charge \( Q \), derivative of admittance \( \partial_A \), and electromotive force \( \mathcal{E} \), respectively), allows one to reinterpret the polarization energy evolutions implied in Fig. 2 as the evolutions of the energies contained in the dispersive elements (inductors and capacitors) of admittance-equivalent circuits. In the case of electrical circuits, the plotted losses correspond to the losses in the dissipative elements of the two circuits, i.e. the losses through the resistors. This time we interpret the discrepancies in these curves as indicating that different LRC circuits with the same admittance can, temporarily, lose energy to their resistors at different rates. Figures 3(b) and 3(d) give LRC circuits corresponding to the mechanical models shown in Figures 3(a) and (c), respectively.

From the discussion above, one concludes that the polarization energy and the associated losses depend intrinsically upon the specific microscopic model giving rise to the susceptibility \( \chi \). However, these allocations cannot be relevant macroscopically: for example, within the phenomenological framework in which \( \chi \) is introduced, the spatial and temporal evolution of the various fields depend only upon this particular piece of information, not upon its various representations. In this circumstance in which the system dynamics is completely determined by some piece of information (e.g. \( \chi \)), to say that some other piece of information is important in order to establish some particular notion of energy allocation is to admit that that notion of energy allocation is irrelevant to those all-ready specified dynamics. We establish a notion of energy allocation that is determined uniquely by \( \chi \) and, so, is relevant to system dynamics. In particular it provides a precise notion of the maximum possibility for the field to recover energy from the medium in, for example, the borrow-return process mentioned at the beginning of this article.

In this article, we introduce the irreversible energy density \( u_{\text{irrev}}(t) \). At any given time \( t \), it is defined to be the infemum of all possible [LL] asymptotic losses \( u_{\text{int}}(+\infty) \), this extrema being realized over all possible future evolutions of the electric field \( E \), holding its past evolution fixed. Temporarily using notation emphasizing that the internal energy at time \( t \) depends not only on this time, but also on the history of the instigating electric field \( E \) up until that time, one writes

\[
u_{\text{irrev}}[E](t) = u_{\text{irrev}}[E^{-}_t](t) := \inf_{E^+_t} [E^-_t + E^+_t](+\infty).
\]

(6)

Here \( E^-_t \) denotes the electric field time series \( E(t) \) with its \( t \)-future \( (\tau > t) \) eliminated. Similarly, \( E^+_t \) denotes an appended electric field time series \( E(t) \) with its \( t \)-past \( (\tau < t) \) eliminated. For a passive dielectric, this infemum exists and, so, is unique \( \chi \). In particular, and is shown later in Eq. (9), it does not depend upon an explicit representation for \( \chi \).

Almost tautologically, from definition (6), it follows that \( u_{\text{irrev}}(t) \) can never decrease as time increases. Thus, at any given time \( t \), it quantifies a component of the medium internal energy \( u_{\text{int}}(t) \) that will, under all circumstances, remain in and be dissipated by the medium. That is it specifies a component of the medium internal energy that cannot under any circumstances be returned to the field. Moreover, since it is defined in terms of an infemum, i.e. a greatest lower bound, all notions of loss greater than this value are too pessimistic: at any given time \( t \), there always exists a future medium-field interaction creating less eventual energy loss to the medium than any value greater than that specified by \( u_{\text{irrev}}(t) \). This is true regardless of how small this overestimation is. Consequently, within the phenomenological, macroscopic framework in which \( \chi \) dictates the system dynamics, this quantity uniquely records the energetic irreversibility generated within this dissipative system.

In Fig. 3 the dot-dashed curve specifies the irreversible energy for the case considered, obviously valid for either of the two \( \chi \)-equivalent physical systems. Note that in the figure it is never exceeded by the losses in the sense of Loudon, Barash and Ginzburg. Indeed one can prove that this relationship must hold between the macroscopically relevant irreversible energy and any notion of loss specified macroscopically. Equivalently one determines that energy (the ability to do work) specified macroscopically cannot exceed any such microscopic notion. Further, one concludes that the former is almost always strictly less than the latter because of incoherence among the system’s microscopic, energy containing elements. The former statement (the one regarding losses) can be obtained by repeated application of the following
theorem: (As in the monotonicity of $u_{\text{irrev}}(t)$, the theorem follows almost tautologically from the definition.)

$$u_{\text{irrev}}[E; \chi_1 + \chi_2](t) \geq u_{\text{irrev}}[E; \chi_1](t) + u_{\text{irrev}}[E; \chi_2](t),$$

with strict inequality holding almost always for non-trivial $\chi_1$ and $\chi_2$. Eq. (3) demonstrates that in "additive" processes (i.e. generating media mixtures), irreversibility is generated. By repeated (additive) subdivisions of a macroscopically relevant susceptibility $\chi$ into pieces $\chi_i$, one may obtain microscopic representations of the medium response. If the elements are "simple" enough (to be quantified later) $u_{\text{irrev}}[E; \chi_i]$ will be equivalent to the parameter-dependent notion of loss suggested by Loudon, and Ginzburg and Barash. Indeed the notion of loss and energy specified by Loudon happen to agree with the macroscopically/phenomenologically relevant notion herein introduced in the cases he considered—single Lorentz oscillator media. [This is not the case for (competing) multiple oscillator media considered by Barash and Ginzburg, as demonstrated by figure 3.]

The difference between the current internal energy and the current irreversible energy gives the reversible energy $u_{\text{rev}}(t)$:

$$u_{\text{rev}}(t) := u_{\text{int}}(t) - u_{\text{irrev}}(t).$$

The reversible energy gives the least upper bound on the amount of energy that the medium can relinquish after time $t$: any amount greater than this value, no matter how small the discrepancy, overestimates the ability of the medium’s microscopic, energy-containing components, say, to organize themselves and do useful, macroscopic work. Obviously the difference between the (piece-wise constant) internal energy plotted in Fig. 3 and the dot-dashed curve in that figure, gives this dynamical notion of the possibility for the medium to do work (against the field) for the example considered.

Using Eq. (4), one immediately shows that $u_{\text{int}}(t)$ is constant after the electric field ceases. From definition (3), and the theorem regarding the monotonicity of $u_{\text{irrev}}(t)$, it follows that $u_{\text{rev}}(t)$ can never increase after such time, i.e. after the electric field quits subsidizing its existence by doing work against the polarized medium. We will show that $u_{\text{rev}}(t)$ is never negative. Consequently, one sees that when the system becomes energetically closed, the reversible energy behaves like a dynamical system free energy (density), equivalently like a system Lyapunov function (density). For this reason, and for the microscopic consideration mentioned, in particular the entropy-generation-like statement embodied in Eq. (5), we will also designate the reversible energy as the (medium-field) free energy.

We finish with a formula demonstrating how the macroscopic loss (and so the free energy (via 5), can be calculated. (In particular how the irreversible energy plotted in figure 3 can be generated.) This formula is obtained by applying a variational principle to the definition (5), and solving the resulting Riemann-Hilbert problem. One finds that, for passive, causal dielectrics,

$$u_{\text{irrev}}[E](t) = \frac{\lambda}{2\pi} \int_{-\infty}^{t} \int_{-\infty}^{+\infty} \frac{e^{-i\omega\tau} E_\tau(\omega)}{\phi_+(\omega)} d\omega d\tau \quad \text{(9)}$$

where

$$\lambda = \lim_{\omega \to \infty} \frac{\text{Im}[\chi(\omega)]}{\omega |\chi(\omega)|^2},$$

$$\phi_+(\omega) = \lim_{\epsilon \to 0^+} \exp \left[ \frac{-1}{2\pi i} \int_{-\infty}^{+\infty} \log \frac{\text{Im}[\chi(\omega)]}{\omega |\chi(\omega)|^2} \frac{d\omega'}{\omega' - (\omega + i\epsilon)} \right] \quad \text{(10)}$$

Here we introduce the instantaneous spectrum at time $t$, $E_\tau(t)$. It is the Fourier transform of $E^{-}_\tau$ (see the lines following definition 3). We also introduce the medium complexity factor $\phi_+(\omega)$. Its deviation from unity gives a measure of the effective macroscopic/phenomenological incoherence of possible microscopic, energy containing elements. Media for which $\phi_+(\omega)$ is identically unity we call simple media, the rest we call complex. In the case that the susceptibility $\chi$ corresponds to a single Lorentz oscillator medium, as in the case considered by Loudon, $\phi_+(\omega)$ is identically one, and Eq. (5) reduces to

$$u_{\text{irrev}}[E](t) = \lambda \int_{-\infty}^{t} \tilde{P}^2(\tau) d\tau \quad \text{(11)}$$

where, then, $\lambda$ is determined in terms of the phenomenological damping parameter $\gamma$ and the effective plasma frequency $\omega_p$:

$$\lambda = \gamma/\omega_p^2. \quad \text{(12)}$$

In such case, then, and as claimed by Loudon, the relevant losses correspond to the frictional losses generated by the single Lorentz oscillator.

**SUMMARY**

We have introduced notions of free energy and losses relevant to the macroscopic behavior of passive, linear dielectric media. These notions are relevant in the same regime in which the macroscopic Maxwell equations are themselves relevant, i.e. in the regime in which that theory specifies all measurable dynamics. In particular, the macroscopic theory introduced is relevant to the regimes in which the macroscopic, energy barrow-return process
describes the production of slow and fast light in passive, linear, temporally dispersive media. Further communications will describe the precise evolutions of the ideal medium-field interactions giving rise to maximum energy recovery, i.e. those "recovery" fields suggested by the variational definition (6). The nature of the analogy of such recovery fields in dissipative media with reversible processes in the thermodynamic setting will be analyzed. Finally, the nonintuitive evolution of the medium-field interaction on such recovery fields in complex media will be exposed, e.g. the failure of monotonicity in the evolution of the internal energy to its minimum value, even on ideal recovery fields, and the identification of such as a measure of the level of macroscopic disorder created by the existence of many, competing degrees of freedom.

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