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Article

Electromagnetic Nanoscale Metrology Based on Entropy Production and Fluctuations *

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Abstract: The goal in this paper is to show how many high-frequency electromagnetic metrology areas can be understood and formulated in terms of entropy evolution, production, and fluctuations. This may be important in nanotechnology where an understanding of fluctuations of thermal and electromagnetic energy and the effects of nonequilibrium are particularly important. The approach used here is based on a new derivation of an entropy evolution equation using an exact Liouville-based statistical-mechanical theory rooted in the Robertson-Zwanzig-Mori formulations. The analysis begins by developing an exact equation for entropy rate in terms of time correlations of the microscopic entropy rate. This equation is an exact fluctuation-dissipation relationship. We then define the entropy and its production for electromagnetic driving, both in the time and frequency domains, and apply this to study dielectric and magnetic material measurements, magnetic relaxation, cavity resonance, noise, measuring Boltzmann's constant, and power measurements.

Keywords: dielectric, entropy; magnetic; nonequilibrium; relaxation.

1. Introduction

The goal of this paper is to systematically analyze dynamically-driven, electromagnetic systems by starting from a detailed microscale theory and then progress to various approximations and show how a wide array of electromagnetic metrology problems relate to entropy, entropy-production rate, and entropy rate fluctuation-dissipation relationships. Since noise and material measurements are now being

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made at the nanoscale, fluctuations and nonequilbrium effects due to temperature gradients and nonuniform fields are becoming increasingly important. Therefore it is important to understand how to model nonequilibrium effects in the measurement process and we will see that entropy production rate and its fluctuations are an important tool. The normal fluctuation-dissipation relations are a special case of the entropy-production, fluctuation-dissipation relationship derived in this paper.

The importance of understanding entropy and its production in relation to metrology has been emphasized and used by a number of researchers [1–3]. Many others have extensively studied the entropy problem [4–7]. In fact some of the most fundamental breakthroughs in modern physics, those of Planck and Einstein for blackbody and brownian motion were originally analyzed by entropy-probability arguments. Electromagnetic measurements of materials and other fundamental properties such as noise and power relate to microscopic quantities that are defined in terms of thermal and electrical correlation functions. The fluctuations originate from random charge motion and thermal fluctuations that produce random polarization, voltage, currents, and magnetic behavior. These fluctuations can be related to dissipation and therefore entropy production.

It is well known that the losses in frequency-dependent electromagnetic material parameters can be determined by the use of weakly applied fields through linear response fluctuation-dissipation relations. The fluctuation-dissipation theorem yields, for example, a relationship between the electric and magnetic susceptibilities and losses[8]. In measurements of high-frequency material properties and noise, the fluctuations in polarizations or voltage are related to the energy dissipated in a material. What is not commonly known is that these types of measurements can also be understood in terms of the entropy-production rate.

The first fluctuation-dissipation relation was derived by Einstein and has been generalized by Kubo [8] and many other researchers. The fluctuation-dissipation relations relate to weak nonequilibrium behavior. In this paper a very general entropy-based form of the fluctuation-dissipation theorem is derived and applied to electromagnetic measurement problems. A linear approximation is obtained and applied to applications in dielectric and magnetic polarization, electrical and magnetic noise, impedance characterization, and resonant phenomena. For this paper we define a system to be in nonequilibrium when there are dynamic applied forces or the system has not had enough time to damp out transient thermal and electrical behavior. Nonequilibrium refer to systems driven by non-time-harmonic external fields or a heat flux across a material or due to relaxation. Specific examples of nonequilibrium electromagnetic driving includes performing measurements of the electric permittivity or permeability when the temperature varies spatially or performing measurements on systems driven by non-time-harmonic fields.

In our analysis the system under study is assumed to consist of a material driven by external fields, which are produced by an external generator. Some of the energy from the driving fields is stored in the fields in the material and some is converted to heat through material relaxation. We begin modeling a system that is closed to the exchange of heat and particles with the outside environment and we later include a heat transfer term in the internal energy.

In this analysis, we concentrate on the projection-operator approach to many-body systems, as developed by Robertson and Zwanzig [9–16]. The projection-operator approach used here has its roots in the work of Zwanzig [10] and Robertson, at the National Bureau of Standards (NBS), and others have

added to its development, for example, Mori, Grabert, and Oppenheim [9–13]. The literature is large and no attempt to overview all of it is made. It has been noted that the projection-operator theory is well-suited for studying entropy production[6, 17]. In this paper we only deal with the projection-operator approach and not with the large literature due to Zubarev's approach or other related approaches[14, 15]. The projection-operator approach defines a nonequilibrium entropy for a dynamically driven system that reduces to the equilibrium entropy in the appropriate limit. The advantage of the projection-operator approach in studying entropy evolution is that the equations a) incorporate both relevant and irrelevant information, b) are exact, Hamiltonian-based, without phase-space compression, c) and are based on reversible equations of motion. The system is described by a set of relevant variables, but there is also assumed to be irrelevant information that is accounted for through the projection operator. This irrelevant correction relates to relaxation and dissipation. The Robertson version of the Zwanzig projection-operator approach obtains a relative canonical density operator by maximizing the entropy subject to constraints on the relevant variables, at given instants in time.

Robertson's exact projection-operator statistical-mechanical theory, which is an exact extension of the Zwanzig and Mori formalism, provides a solid basis for analysis of electromagnetic driving[9, 18]. The theory can be formulated either quantum-mechanically or classically. Oppenheim and Levine extended Robertson's work, included a more general initial condition, and studied a linear approximation to entropy evolution [13]. Projection-operator approaches have been used to study the microscopic time evolution of electromagnetic properties [19–21]. In Robertson's approach the full density operator $\rho(t)$ equals a relevant canonical density operator $\sigma(t)$ plus a relaxation correction term that accounts for irrelevant information. In this exact, time-symmetric formulation, the statistical density operator satisfies the Liouville equation, whereas the relevant canonical-density operator does not. This theory yields an expression that exhibits all the required properties of a nonequilibrium entropy, and yet it is based entirely on time-symmetric equations.

It is well-known that the maximum of the information-theoretical entropy subject to a set of constraints has maximal uncertainty [22–24]. This entropy is not the full nonequilibrium entropy because it does not contain information on the irrelevant variables. In the projection-operator approach used in this paper, a Gibbs-like relevant canonical-density operator $\sigma(t)$ is constructed by maximizing the entropy subject to relevant constraint information. This projection-operator approach, developed by Robertson [9], marries the Jaynes information-theoretic approach to the exact solution of the equations of motion by accounting for both relevant and irrelevant information. It is this correction term that relates to relaxation and is absent in the Jaynes information-theoretic and Gibbsian approaches. Since the Robertson approach is exact, and includes a correction for all irrelevant information when determining the density function, it is equivalent to a limit of a Jaynesian expression for a probability density that contains all possible information.

This paper starts by reviewing, from a very basic starting point, the statistical-mechanical foundations of entropy-production rate from a projection-operator approach. A new, very general and exact relation is derived for the entropy rate in terms of fluctuations in the microscopic entropy, then an exact entropy-density equation is developed that includes entropy flux and production. We then apply this entropy expression in various linear approximations to describe electromagnetic applications. These results are

then used to show that many areas of electromagnetic metrology can be based on the time-dependent and frequency-dependent entropy concept where polarization, voltage, and current fluctuations are seen as specific components. Applications are made to electromagnetic material measurements, noise, cavity resonance, and relaxation. The fundamental results derived here are valid both at equilibrium or far from thermal or electrodynamic equilibrium and form a basis for extending measurements into the nanoscale and nonequilibrium realms.

2. Theoretical Analysis of the Entropy

2.1. Background

In this section the formalism and equations are derived for the entropy and its related evolution under electromagnetic driving. We know that changes in entropy satisfy $\delta S = \delta Q/T + \delta S_i$, where δQ is the generalized heat added to the system, such as heat flowing in through the system boundaries and δS_i is the entropy change due to irreversible processes such as relaxation. For a closed system, $\delta Q = 0$ and $\delta S = \delta S_i \geq 0$ [5, 25–28]. As the system is dynamically driven by applied fields, the material relaxes and local fields are formed in the material that differ from the applied fields. As a consequence, a new energy configuration is formed. The origin of relaxation is the process of transforming from applied fields acting on the material to local fields acting on materials.

The dynamical variables we use are a set of operators, or classically, a set of functions of phase $F_1(\mathbf{r}), F_2(\mathbf{r}), \cdots$. For normalization, $F_0 = 1$ is included in the set. These operators are, for example, the internal-energy density $u(\mathbf{r})$, and the electromagnetic polarizations $\mathbf{m}(\mathbf{r})$, $\mathbf{p}(\mathbf{r})$. The operators F_n are functions of $\mathbf{r}$ and phase variables, but are not explicitly time dependent. The time dependence enters when the trace is taken, through the driving fields and in the Hamiltonian. Associated with these operators are a set of thermodynamic fields, that are not operators and do not depend on phase, such as generalized temperature and local electromagnetic fields such as $\mathbf{E}_p$ and $\mathbf{H}_m$ and temperature. In any complex system, in addition to the set of F_n, there are many other uncontrolled or unobserved variables that are categorized as irrelevant variables.

A brief overview of the approach for calculating the equations of motion for the relevant variables will now be presented, for details please refer to [6, 9, 19]. Later, these results will be used to derive an entropy-fluctuation relation. There are two density operators. The first is the full statistical-density operator $\rho(t)$ that satisfies the Liouville equation

$$d\rho/dt = -i\mathcal{L}\rho = \frac{1}{i\hbar}[\mathcal{H}(t), \rho]. \tag{1}$$

Note we are using the Fourier transform time convention $e^{-i\omega t}$, $\mathcal{L}(t)$ is the time-dependent Liouville operator, and $\mathcal{H}(t)$ is the Hamiltonian that is time dependent because the applied fields are time dependent.

In addition to ρ we define the relevant canonical-density operator $\sigma(t)$ that is developed by maximizing the information entropy subject to constraints on the expected values of operators.

The entropy is defined as

$$S(t) = -k_B Tr\left(\sigma(t) \ln \sigma(t)\right), \tag{2}$$

where Tr denotes trace. In Eq.(2), σ is formed from the relevant variables and is constructed through maximizing the entropy subject to constraints on the expectations of the operators $< F_n(\mathbf{r}) >= Tr(F_n(\mathbf{r})\sigma(t))$. Maximization by the common variational procedure leads to the generalized canonical density,

$$\sigma(t) = \exp\left(-\lambda(t) * F\right), \tag{3}$$

where we require

$$Tr(\exp\left(-\lambda(t) * F\right)) = 1. \tag{4}$$

In Eq.(4), $\lambda(\mathbf{r}, t)$ are Lagrangian multipliers that correspond to local nonquantized fields, such as temperature and electromagnetic fields. We use the notation $\lambda * F = \int d^3r \sum_n \lambda_n(r,t)F_n(r)$. The constraints for $n = 0, 1, 2, 3...$ requires that the expected values of the relevant variables, (but not their derivatives, etc.), with respect to σ and ρ are equal at each time:

$$Tr(F_n(\mathbf{r})\rho(t)) \equiv Tr(F_n(\mathbf{r})\sigma(t)) \equiv< F_n >= Tr\left(F_n \exp\left(-\lambda(t) * F\right)\right). \tag{5}$$

In our analysis the relevant variables will be the polarizations and the internal energy.

The dynamical evolution of the relevant variables, that is, the evolution from the Hamiltonian, is described by

$$\dot{F}_n(\mathbf{r}) = i\mathcal{L}F_n(\mathbf{r}) = -\frac{1}{i\hbar}[\mathcal{H}(t), F_n(\mathbf{r})], \tag{6}$$

where the Hamiltonian is, for example, in electromagnetic driving: $\mathcal{H}(t) = \int (u(\mathbf{r}) - \mathbf{p}(\mathbf{r}) \cdot \mathbf{E}(\mathbf{r}, t) - \mu_0 \mathbf{m}(\mathbf{r}) \cdot \mathbf{H}(\mathbf{r}, t))d^3r$. In addition to the dynamical evolution, we will see that there are also changes in the relevant variables evolution due to the irrelevant information. An important identity was proven previously [18]

$$i\mathcal{L}\sigma(t) = -\lambda * \overline{\dot{F}}\sigma, \tag{7}$$

where the bar is defined for any operator A as $\overline{A} = \int_0^1 \sigma^x(t) A \sigma^{-x}(t) dx$. In a classical analysis $\overline{A} = A$. Also $Tr(i\mathcal{L}\sigma(t)) = 0$.

Robertson developed an exact equation for $\rho(t)$, that contains memory, in terms of $\sigma(t)$. When solving that equation and using Oppenheim's extended initial condition we obtain [13]

$$\rho(t) = \sigma(t) + \mathcal{T}(t, 0)\chi(0) - \int_0^t d\tau \mathcal{T}(t, \tau)\{1 - P(\tau)\} i\mathcal{L}(\tau)\sigma(\tau), \tag{8}$$

for the initial condition $\chi(0) = \rho(0) - \sigma(0)$ (note that Oppenheim and Levine [13] generalized the analysis of Robertson to include this more generalized initial condition). $\mathcal{T}$ is an evolution operator, $\mathcal{T}(t, t) = 1$, and satisfies

$$\frac{\partial \mathcal{T}(t, \tau)}{\partial \tau} = \mathcal{T}(t, \tau)(1 - P(\tau))i\mathcal{L}(\tau), \tag{9}$$

where P is a nonhermitian projection-like operator defined by the functional derivative

$$P(t)A \equiv \sum_{n=1}^{m} \int d^3r \frac{\delta\sigma(t)}{\delta < F_n(\mathbf{r}) >} Tr(F_n(\mathbf{r})A), \tag{10}$$

for any operator A [9]. In Robertson's pioneering work he has shown that Eq.(10) is equivalent to the Kawasaki-Grunton and Grabert's projection operators, and is a generalization of the Mori and Zwanzig

projection operators[29]. As a consequence of this definition of the projection operator: $\partial\sigma/\partial t = P\partial\rho/\partial t$. The normal maximum entropy procedure (MAXENT) neglects the last term on the RHS of Eq.(8).

For the very special, but unrealistic case, when all $\dot{F}_n(\mathbf{r})$ can be written as a linear sum of the set $F_m(\mathbf{r})$

$$\dot{F}_n(\mathbf{r}) = \sum_m \int d^3r' a_m(\mathbf{r},\mathbf{r}')F_m(\mathbf{r}'), \tag{11}$$

for all n. If one could write this for all n then the last terms on the RHS of Eq.(8) are zero and then $\sigma(t) = \rho(t)$: in general $\rho(t) \neq \sigma(t)$. Here the function a_m does not depend on the phase variables. This is a consequence of the identity $PF_m\sigma = F_m\sigma$ [13, 29]. When Eq.(11) applies for all n(see Robertson [21] Eq.(A7) Oppenheim and Levine [13]), the relaxation terms in Eq.(8) are absent. Eq.(11) would not apply for any macroscopic system and in addition causality, through the Kramer's-Kronig condition, requires dissipation. As noted by Oppenheim and Levine and Robertson[9, 13], it is the failure of Eq.(11) for almost all physical systems, that produces the relaxation term in Eq.(8) and the resultant entropy production. Due to the invariance of the trace operation under unitary transformations, it is known that the Neumann entropy $-k_B Tr(\rho(t)\ln\rho(t))$ formed from the full statistical-density operator $\rho(t)$, that satisfies the Liouville equation, is independent of time. This is very seldom realized in measurement systems where irrelevant variables influence the system's evolution.

In the above, we have only considered closed, dynamically driven systems. In an open system, $\rho(t)$ may not evolve unitarily and $\rho(t)$ need not satisfy Eq.(1)[16].

It has been shown that the exact time evolution of the relevant variables can be expressed for a dynamically-driven system as [9, 13]

$$\frac{\partial < F_n(\mathbf{r}) >}{\partial t} =< \dot{F}_n(\mathbf{r}) > + Tr(\dot{F}_n(\mathbf{r})\mathcal{T}(t,0)\chi(0)) - \int_0^t Tr\left(i\mathcal{L}F_n(\mathbf{r})\mathcal{T}(t,\tau)(1-P(\tau))i\mathcal{L}\sigma(\tau)\right)d\tau. \tag{12}$$

Equations (5) and (12) form a closed system of equations and the procedure for determining the Lagrange multipliers in terms of $< F_n >$ is to solve Eqs.(5) and Eq.(12) simultaneously. For operators that are odd under time reversal, such as the magnetic moment, the first term on the right hand side of Eq.(12) is nonzero, whereas for functions even under time reversal, such as dielectric polarization, and microscopic entropy, this term is zero. However, the third term in Eq.(12) in any dissipative system is nonzero. The relaxation correction term that appears in the projection-operator formalism is essential and is a source of the time-dependence in the entropy rate. Although these equations are nonlinear in many cases linear approximations have been successfully made[30]. For open systems Eq.(12) is modified only by adding a source term[31].

When $\dot{F}_n(\mathbf{r}) = -\nabla\cdot\mathbf{j}_m(\mathbf{r})$ and $\nabla\cdot\mathbf{J}_m(\mathbf{r},t) = -\partial < F_m(\mathbf{r}) > /\partial t$, we can express Eq.(12) using integration by parts in terms of currents[18]

$$\mathbf{J}_m(\mathbf{r},t) =< \mathbf{j}_m(\mathbf{r}) > + \sum_n \int_0^t \int d\mathbf{r}' \underbrace{Tr\left(\frac{\mathbf{j}_m(\mathbf{r},t)\mathcal{T}(t,\tau)(1-P(\tau))\bar{\mathbf{j}}_n(\mathbf{r}',\tau)}{E_\beta(\mathbf{r}',\tau)}\sigma(\tau)\right)}_{\overleftrightarrow{K}_{mn}} \cdot\nabla\lambda_n(\mathbf{r}',\tau)d\tau. \tag{13}$$

The mean energy of a quantum oscillator defines the temperature $E_\beta = (\hbar\omega/2)\coth(\hbar\omega/2k_BT)$ and in a high temperature approximation this reduces to $E_\beta \to k_BT$. Approximate transport coefficients and the related fluctuation-dissipation relations for conductivity, susceptibility, noise, and other quantities follow naturally from Eq.(13), if we have time-harmonic fields, and take a linear, time-invariant approximation. If we take the Laplace transform (L), then, $\widetilde{\mathbf{J}}_m(\mathbf{r},\omega) = \sum_n \overleftrightarrow{\sigma}_{mn}(\mathbf{r},\omega)\cdot\widetilde{\mathbf{E}}(\mathbf{r},\omega)$ where

$$\overleftrightarrow{\sigma}_{mn}(\mathbf{r},\omega) = \int d\mathbf{r}' L\left[\overleftrightarrow{K}_{mn}\right]. \tag{14}$$

We will use Eq.(14) in our applications to electromagnetic constitutive properties.

2.2. *The Entropy in the Projection-Operator Formulation*

From Eq.(2) and using Eq.(3) we define the entropy for a set of relevant variables: the electric polarization, $\mathbf{P} = Tr(\mathbf{p}\sigma)$, the magnetic polarization, $\mathbf{M} = Tr(\mathbf{m}\sigma)$, and the internal-energy density, $U = Tr(\mathbf{u}\sigma)$

$$\begin{aligned} S(t) &= -k_B < \ln(\sigma) >= k_B\lambda(\mathbf{r},t) * < F(\mathbf{r}) > \\ &= k_B \int \left[\frac{U - \mathbf{P}\cdot\mathbf{E}_p - \mu_0\mathbf{M}\cdot\mathbf{H}_m + E_\beta \ln Z}{E_\beta}\right] d^3r. \end{aligned} \tag{15}$$

$F_0 = 1$ has been included in the set of operators for normalization. The free energy is $\mathcal{F} = -k_BT\ln Z$. The Lagrangian multipliers in this example were $-\mathbf{E}_p/E_\beta \to -\mathbf{E}_p/k_BT$, $-\mathbf{H}_p/E_\beta \to -\mathbf{H}_m/k_BT$, and $1/E_\beta \to 1/k_BT$.

In the following sections we will apply the theory we have developed to various problems in electromagnetism. Before doing this, we need to define the entropy density and production rate. The entropy density $s_d(t)$ follows a conservation equation of the form

$$\frac{\partial s_d(\mathbf{r},t)}{\partial t} + \nabla\cdot\mathbf{J}_e(\mathbf{r},t) = \Sigma_d(\mathbf{r},t), \tag{16}$$

where $\mathbf{J}_e$ is an entropy current; for example, due to a heat flux flowing into the system. In Eq.(16), Σ_d is the entropy density production rate due to irreversible processes and relaxation. The units of Σ_d are entropy per second per unit volume.

The total microscopic entropy production rate, which is the integrated-entropy density production rate, originates from the dynamical evolution of the relevant variables We define the total microscopic entropy production rate as

$$\dot{s}(t) \equiv k_B\lambda * \dot{F} = -k_B\lambda * i\mathcal{L}F. \tag{17}$$

The expected value of the dynamical contribution to the total entropy production rate vanishes due to Eq. (7)[13]:

$$< \dot{s}(t) >= Tr(\dot{s}(t)\sigma) = k_BTr(\lambda * \dot{F}\sigma) = k_B\lambda * Tr(\dot{F}\sigma) = 0. \tag{18}$$

This result follows from time-reversal invariance of the trace of $Tr(i\mathcal{L}\sigma)$, using the cyclic invariance of the trace, and Eq.(6). Equation (18) is a result of the microreversiblity of the equations of motion of the relevant variables. For example, in the case of an isolated system with dynamical electromagnetic driving

that has microscopic internal energy u, magnetization $\mathbf{m}$, local field $\mathbf{H}_m$, and generalized temperature T, we would have from Eq.(18): $k_B \int d^3r\{< \dot{u}(\mathbf{r}) > -\mu_0 < \dot{\mathbf{m}}(\mathbf{r}) > \cdot\mathbf{H}_m(\mathbf{r},t))/E_\beta(\mathbf{r},t)\} = 0$. In other words, in the dynamical contribution to the evolution of the relevant variables, all contributions to the entropy rate are taken into account and in this sense $< \dot{s} >$ does not directly influence the total entropy evolution dS/dt, which includes the effects of both the relevant and nonrelevant variables in the dissipative term.

The total entropy evolution can be formed from Eq.(12) by multiplying by λ and integration over space

$$\begin{aligned}\frac{dS(t)}{dt} &\equiv k_B\lambda * \frac{\partial < F >}{\partial t} = Tr(\chi(0)\dot{s}(t)\mathcal{T}(t,0)) \\ &+ \int_0^t Tr\left(i\mathcal{L}s(t)\mathcal{T}(t,\tau)(1-P(\tau))i\mathcal{L}\sigma(\tau)\right)d\tau = Tr(\rho(0)\dot{s}(t)\mathcal{T}(t,0)) \\ &+Tr(\dot{s}(t)(\sigma(t)-\sigma(0)\mathcal{T}(t,0))) + \frac{1}{k_B}\int_0^t Tr\left(\dot{s}(t)\mathcal{T}(t,\tau)(1-P(\tau))\bar{\dot{s}}(\tau)\sigma(\tau)\right)d\tau.\end{aligned} \tag{19}$$

Equation (19) is an exact expression of the second law of thermodynamics since the Robertson-Zwanzig statistical-mechanical theory is an exact quantum-mechanical solution of the Liouville equation applied to the relevant variables, without approximation (see Robertson [18]). Note that it is time-reversal invariant, but also models dissipation. We used the relation $< \dot{s}(t) >= 0$. The last expression in Eq.(19) indicates that the entropy production rate satisfies a fluctuation-dissipation relationship in terms of the microscopic entropy production rate$\dot{s}(t) = i\mathcal{L}s(t)$. At $t = 0$, $dS/dt(t = 0) = Tr(\rho(0)\dot{s}(0))$. Equation(19) will form the basis of our applications to various electromagnetic driving and measurement problems. The LHS of Eq.(19) represents the dissipation and the last term on the RHS represents the fluctuations in terms of the microscopic entropy production rate $\dot{s}(t)$. Due to incomplete information there are contributions from the positive semi-definite relaxation terms in Eq.(19) for almost all many-body systems. For a dynamically-driven system $dS/dt - Tr(\chi(0)\dot{s}(t)\mathcal{T}(t,0)) \geq 0$. For an open system Eq.(19) would be modified by adding an entropy source term. To summarize, for a dynamically driven system, the expected value of the microscopic entropy production rate is zero due to the microscopic reversibility of the underlying equations of motion; however in a complex system there are other uncontrolled variables in addition to the relevant ones that act to produce dissipation and irreversibility and a net positive macroscopic entropy evolution. Since this equation is exact, systems away from equilibrium can be modeled.

Equation (19) could be used to determine Boltzmann's constant if dS/dt was obtained from a measurement of the losses in a system and the trace expression was determined by measurements of the fluctuations of the entropy production. This could be actualized, in principle, by the measurement of noise in an electrical system, similar to the studies of determining Boltzmann's constant from Johnson noise. It seems intuitive that Boltzmann's constant could be determined by entropy measurements, since they both have the same units.

For a stationary process, the RHS of Eq. (19) can be expressed through the Wiener-Khinchine theorem, over a bandwidth of Δf, and cast into a form that relates the net dissipative entropy production, av-

eraged over a cycle, in terms of entropy-production fluctuations. A special case of this equation is Johnson noise where $dS/dt \rightarrow (1/2)I_0^2 R$ and $< \dot{s}_\omega^2 >_0 \rightarrow (1/4)I_0^2 < v^2 >_0$, that yields $4k_B RT\Delta f =< v^2 >$. The entropy-production correlation function can be measured in terms of power fluctuations. We will use this relation for applications in electromagnetism and indicate how it relates to the fluctuation-dissipation theorems for dielectric and magnetic measurements, Johnson noise, and Boltzmann's constant determination.

Using Eq.(19), $\dot{s}(t) = k_B \lambda * \dot{F}$, and assuming a continuity equation for the evolution, the conserved relevant variables can be written as $\dot{F}_m(\mathbf{r}) = -\nabla \cdot \mathbf{j}_m(\mathbf{r})$, where the normal component of $\mathbf{j}_m$ vanishes on bounding surfaces, and the rest are nonconserved variables. The entropy-density balance equation is

$$\frac{\partial s_d(\mathbf{r},t)}{\partial t} + \nabla \cdot \underbrace{\int_0^t \sum_{m(cons)} Tr\left(\lambda_m(\mathbf{r},t) \cdot \mathbf{j}_m(\mathbf{r},t)\mathcal{T}(t,\tau)(1-P(\tau))\bar{\dot{s}}(\tau)\sigma(\tau)\right) d\tau}_{\mathbf{J}_e}$$

$$= \int_0^t \sum_{m(cons)} Tr\left(\nabla\lambda_m(\mathbf{r},t) \cdot \mathbf{j}_m(\mathbf{r},t)\mathcal{T}(t,\tau)(1-P(\tau))\bar{\dot{s}}(\tau)\sigma(\tau)\right) d\tau$$

$$+ \int_0^t \sum_{n(non)} Tr\left(\lambda_n(\mathbf{r},t) \cdot \dot{F}_n(\mathbf{r})\mathcal{T}(t,\tau)(1-P(\tau))\bar{\dot{s}}(\tau)\sigma(\tau)\right) d\tau. \tag{20}$$

For an open system entropy fluxes may travel through the boundaries and may be modeled by the addition of an entropy source.

Equation (19) can also be used as a generator of the relaxation part of the equations of motion of the relevant variables. Taking a functional derivative of Eq.(19) w.r.t. λ_k, and noting that the volume integral over $\mathbf{r}$ can be arbitrary, we obtain the exact evolution equations, without the reversible contribution, that were derived by Robertson and later by Oppenheim [9, 13] and others, given in Eq.(12). For example, if the driving forces are the related Lagrange multipliers, $-\mathbf{E}_p/k_BT$, $-\mathbf{H}_m/k_BT$, and $1/k_BT$, we obtain the equation of motion of the polarization response $\partial\mathbf{P}/\partial t$, (see Baker-Jarvis et al. [6, 20]):

$$\frac{\partial \mathbf{P}(\mathbf{r},t)}{\partial t} = Tr(\chi(0)\dot{\mathbf{p}}(\mathbf{r})\mathcal{T}(t,0)) - \int_0^t Tr\left(\dot{\mathbf{p}}(\mathbf{r})\mathcal{T}(t,\tau)(1-P(\tau))i\mathcal{L}\sigma(\tau)\right) d\tau$$

$$= Tr(\chi(0)\dot{\mathbf{p}}(\mathbf{r})\mathcal{T}(t,0)) + \frac{1}{k_B}\int_0^t Tr\left(\dot{\mathbf{p}}(\mathbf{r})\mathcal{T}(t,\tau)(1-P(\tau))\bar{\dot{s}}(\tau)\sigma(\tau)\right) d\tau. \tag{21}$$

The second form in Eq.(21) displays the interaction with the microscopic entropy production rate. As another illustration, if we vary the generalized force $-\mathbf{H}_m/T$ in Eq.(19) we obtain the equation of motion for the magnetization

$$\frac{\partial \mathbf{M}(\mathbf{r},t)}{\partial t} = Tr(\chi(0)\dot{\mathbf{m}}(\mathbf{r})\mathcal{T}(t,0)) - \int_0^t Tr\left(\dot{\mathbf{m}}(\mathbf{r})\mathcal{T}(t,\tau)(1-P(\tau))i\mathcal{L}\sigma(\tau)\right) d\tau$$

$$=< \dot{\mathbf{m}}(\mathbf{r}) > + Tr(\chi(0)\dot{\mathbf{m}}(\mathbf{r})\mathcal{T}(t,0)) + \frac{1}{k_B}\int_0^t Tr\left(\dot{\mathbf{m}}(\mathbf{r})\mathcal{T}(t,\tau)(1-P(\tau))\bar{\dot{s}}(\tau)\sigma(\tau)\right) d\tau. \tag{22}$$

We can identify $\dot{\mathbf{m}} = -\mu_0|\gamma_{eff}|\mathbf{m} \times \mathbf{H}_{eff}$ where γ_{eff} is the effective gyromagnetic factor. The equation

of motion for the internal-energy density is

$$\frac{\partial U(\mathbf{r},t)}{\partial t} = Tr(\chi(0)\dot{u}(\mathbf{r})\mathcal{T}(t,0)) - \int_0^t Tr\left(\dot{u}(\mathbf{r})\mathcal{T}(t,\tau)(1-P(\tau))i\mathcal{L}\sigma(\tau)\right)d\tau$$
$$=< \dot{u}(\mathbf{r}) > + Tr(\chi(0)\dot{u}(\mathbf{r})\mathcal{T}(t,0)) + \frac{1}{k_B}\int_0^t Tr\left(\dot{u}(\mathbf{r})\mathcal{T}(t,\tau)(1-P(\tau))\bar{\dot{s}}(\tau)\sigma(\tau)\right)d\tau. \quad (23)$$

Equations (21) through (23) are exact, coupled nonlinear equations that must be solved in conjunction with Eqs. (5) for the unknowns: λ_n and $< F_n >$. In general, this is not a simple task, but in many examples it is possible to make approximations to linearize the kernel. Robertson showed how Eq.(22) reduces to the Landau-Lifshitz equation for appropriate assumptions when the kernel $\overleftrightarrow{K}$ was approximated by using $\mathbf{M}\times(\mathbf{M}\times\mathbf{H})\delta(t-\tau) = (\mathbf{MM} - |\mathbf{M}|^2\overleftrightarrow{I})\cdot(\mathbf{H}-\mathbf{H}_m)\delta(t-\tau) \propto \overleftrightarrow{K}\cdot(\mathbf{H}-\mathbf{H}_m)$. The electric polarization Eq.(21) was linearized and solved in [30].

3. Applications to Electromagnetic Driving

In the previous sections we developed the formalism that describes entropy production rate from a projection-operator perspective. We now turn to applications of entropy production rate in electromagnetic driving. Since the exact kernels in Eqs.(21) through (23) are very complicated nonlinear functions of the Lagrange multipliers, to achieve progress, we make linear approximations that maintain the form of the equations as we done previously for other applications[30]. The total entropy production rate for isolated systems driven by electromagnetic fields can be written from Eq.(19) as

$$\frac{dS(t)}{dt} = \int d^3r \frac{1}{T}\left[\frac{\partial U}{\partial t} - \frac{\partial \mathbf{P}}{\partial t}\cdot\mathbf{E}_p - \mu_0\frac{\partial \mathbf{M}}{\partial t}\cdot\mathbf{H}_m\right], \quad (24)$$

where the evolution terms each satisfy Eqs.(21) through (23) and $\mathbf{E}_p$ and $\mathbf{H}_m$ are the Lagrangian multiplier, effective fields. Energy conservation requires

$$\frac{\partial U(\mathbf{r},t)}{\partial t} = \mu_0\frac{\partial \mathbf{M}(\mathbf{r},t)}{\partial t}\cdot\mathbf{H}(\mathbf{r},t) + \frac{\partial \mathbf{P}(\mathbf{r},t)}{\partial t}\cdot\mathbf{E}(\mathbf{r},t). \quad (25)$$

This equation can be used in Eq.(24).

3.1. Entropy Evolution and Production in Magnetic Relaxation

a) As an example, we study entropy production rate from the loss in ferromagnetic materials when modeled by the Landau-Lifshitz approximation to the relaxation term in Eq.(22). We use $\partial s_d/\partial t = \{\partial u/\partial t - \partial \mathbf{M}/\partial t\cdot\mathbf{H}_m\}/T$, the conservation of energy condition: $\partial u/\partial t = (\partial\mathbf{M}/\partial t)\cdot\mathbf{H}$, and the definition $(\partial\mathbf{M}/\partial t)\cdot(\mathbf{H}-\mathbf{H}_m) \equiv (\partial\mathbf{M}/\partial t)\cdot\Delta\mathbf{H} \equiv (\partial\mathbf{M}/\partial t)\cdot\mathbf{H}_{eff}$, where $\Delta\mathbf{H} = \mathbf{H}-\mathbf{H}_m$, and we use the commutator relationship $[(u-\mu_0\mathbf{m}\cdot\mathbf{H}_m),\sigma] = 0$ to eliminate $[u,\sigma]$ in $[\mathcal{H}(t),\sigma]$. The entropy production rate due to magnetic-field driving of a isothermal ferromagnetic material where the loss is modeled by the Landau-Lifshitz loss expression, that is an approximation to the integral term in Eq.(22) $(-\mu_0\gamma_e\alpha/|\mathbf{M}_s|)\mathbf{M}\times(\mathbf{M}\times\mathbf{H}_{eff})$, where γ_e is the gyromagnetic ratio, α is the Landau-Lifshitz

dissipation constant, and $\mathbf{M}_s$ is the saturization magnetization[32]

$$\Sigma_d(\mathbf{r},t) = \frac{\partial \mathbf{M}(\mathbf{r},t)}{\partial t}\cdot\frac{\mathbf{H}(\mathbf{r},t)-\mathbf{H}_m(\mathbf{r},t)}{T} \approx k_B\frac{\mu_0|\gamma_e|\alpha}{|\mathbf{M}_s|}\frac{(\mathbf{M}(\mathbf{r},t)\times\mathbf{H}_{eff}(\mathbf{r},t))\cdot(\mathbf{M}(\mathbf{r},t)\times\mathbf{H}_{eff}(\mathbf{r},t))}{k_BT}. \quad (26)$$

b) As another example that has been studied by Slonczewski[33] and many others, consider the spin-torque transfer due to a polarized electron current from a reference layer, that travels through a free layer, and enters a thin ferromagnetic material that has magnetization $\mathbf{M}$ and applied field $\mathbf{H}$. In this case the spin-current is a source term in an open system in Eq.(22)[31]. The Hamiltonian is $\mathcal{H}(t) \rightarrow \int d^3r\,(u-\mu_0\mathbf{m}\cdot\mathbf{H})$ and $\mathcal{U} = \int u(r)d^3r$. Therefore we can write Eq.(22), with a spin current source term $-\nabla\cdot\overleftrightarrow{\mathbf{Q}}_s$, as:

$$\frac{\partial \mathbf{M}(\mathbf{r},t)}{\partial t} \approx -\mu_0|\gamma_e|\mathbf{M}(\mathbf{r},t)\times(\mathbf{H}(\mathbf{r},t)-\mathbf{H}_m(\mathbf{r},t)) -\nabla\cdot\overleftrightarrow{\mathbf{Q}}_s + \frac{1}{\hbar^2}\int_0^t\int d^3r'Tr\left([\mu_0\mathbf{m}(\mathbf{r}),\mathcal{U}(\mathbf{r})]\mathcal{T}(t,\tau)(1-P(\tau))[\mathbf{m}(\mathbf{r}),\sigma]\cdot(\mathbf{H}-\mathbf{H}_m)\right)d\tau. \quad (27)$$

If we use the Landau-Lifshitz approximation for the relaxation term [21] we obtain

$$\frac{\partial \mathbf{M}(\mathbf{r},t)}{\partial t} = -\mu_0|\gamma_e|\mathbf{M}\times\mathbf{H}_{eff} - \frac{\mu_0|\gamma_e|\alpha}{|\mathbf{M}_s|}\mathbf{M}\times(\mathbf{M}\times\mathbf{H}_{eff}) - \nabla\cdot\overleftrightarrow{\mathbf{Q}}_s\,. \quad (28)$$

As an approximation, the Slonczewski expression for spin-torque transfer can be identified with the spin-source term $-\nabla\cdot\overleftrightarrow{\mathbf{Q}}_s = c\mathbf{M}\times(\mathbf{M}\times\mathbf{M}_e)$, where c is a constant.

The associated total entropy production rate can be obtained using the conservation of energy relationship $\partial U/\partial t = \partial\mathbf{M}/\partial t\cdot\mathbf{H}$ in Eq.(24)

$$\frac{dS}{dt} = \int d^3r\frac{1}{T}\left(\frac{\partial \mathbf{M}(\mathbf{r},t)}{\partial t}\cdot(\mathbf{H}-\mathbf{H}_m)\right), \quad (29)$$

or in analogy to Eq. (16) and using Eq.(28)

$$\frac{ds_d}{dt} + \nabla\cdot\left(\frac{\overleftrightarrow{\mathbf{Q}}_s\cdot\Delta\mathbf{H}}{T}\right) = \frac{\mu_0|\gamma_e|\alpha}{T|M_s|}(\mathbf{M}\times\mathbf{H}_{eff})\cdot(\mathbf{M}\times\mathbf{H}_{eff}) + \frac{\nabla T\cdot\overleftrightarrow{\mathbf{Q}}_s\cdot\Delta\mathbf{H}}{T^2} - \frac{\overleftrightarrow{\mathbf{Q}}_s:\nabla(\Delta\mathbf{H})}{T}. \quad (30)$$

3.2. Entropy Evolution and Production From Electromagnetic Driving

Consider a material that is driven by electromagnetic fields, which now may exchange heat with the surroundings. We assume that the material contains permanent electric and magnetic dipoles. The field power transmitted into the material is partitioned into the internal energy stored in the fields, the energy dissipated in material losses, the work performed by the fields to polarize dielectric or magnetic material, and the energy to drive currents on any conductors in the system. Losses in materials and

currents transform field energy into heat energy. For example, the dissipative currents on cavity walls are formed from the conversion of some of the electromagnetic energy that enters the cavity into mechanical motion of conduction electrons and heat. The transformation of electromagnetic field energy into the kinetic energy of the currents on the cavity walls and in the material results in entropy production. The origin of the entropy production rate is from the work done by an external electromagnetic generator that maintains the fields in the cavity.

The applied electromagnetic fields are $\mathbf{E}$ and $\mathbf{H}$, and the local fields in the material are $\mathbf{E}_p$, and $\mathbf{H}_m$. The power density applied to the system by the remote generator is $-\nabla\cdot(\mathbf{E}\times\mathbf{H})$, which, for an impedance-matched system, adds internal energy to the system at a rate of $-\int\nabla\cdot(\mathbf{E}\times\mathbf{H})dV = -\int\mathbf{E}\times\mathbf{H}\cdot d\mathbf{S}$, where the surface of integration includes the cross-sectional area of the input and output ports and the surface normal is outward. The Poynting vector of the dynamically driven fields together with temperture gradients produce the nonequilibrium in the system. There also may be heat exchange with a reservoir that yields a heat flux $-\nabla\cdot\mathbf{Q}_h$. The Hamiltonian is $\mathcal{H}(t)=\int\{u(\mathbf{r})-\mathbf{p}(\mathbf{r})\cdot\mathbf{E}(\mathbf{r},t)-\mathbf{m}(\mathbf{r})\cdot\mathbf{H}(\mathbf{r},t)\}d^3\mathbf{r}$. The internal energy contains the kinetic energy, electrostatic potential energy, dipole-dipole, spin-spin, spin-lattice, and other interactions. As the applied field impinges on a material specimen, depolarization fields may be formed in the material that modify the fields in the material ($\mathbf{E}_p,\mathbf{H}_m$) from the applied field.

In the analysis, we will need expressions for the rate of change of the stored electromagnetic energy averaged over a cycle. Using Eqs. (21) and (22) we can write

$$\begin{aligned}&\frac{\partial\mathbf{D}(\mathbf{r},t)}{\partial t}\cdot\mathbf{E}_p(\mathbf{r},t)+\frac{\partial\mathbf{B}(\mathbf{r},t)}{\partial t}\cdot\mathbf{H}_m(\mathbf{r},t)\\&\quad=\frac{1}{k_B}\int_0^t\int d^3r'Tr\left((\mathbf{E}_p(\mathbf{r},t)\cdot\dot{\mathbf{p}}(\mathbf{r})+\mu_0\mathbf{H}_m(\mathbf{r},t)\cdot\dot{\mathbf{m}}(\mathbf{r}))\mathcal{T}(t,\tau)(1-P(\tau))\bar{\dot{s}}(\tau)\right)d\tau\\&\quad+\frac{\epsilon_0}{2}\frac{\partial|\mathbf{E}_p|^2}{\partial t}+\frac{\mu_0}{2}\frac{\partial|\mathbf{H}_m|^2}{\partial t}\approx-\nabla\cdot(\mathbf{E}_p\times\mathbf{H}_m)-2\omega_0\epsilon''(\omega_0)\overline{\mathbf{E}_p^2}-2\omega_0\mu''(\omega_0)\overline{\mathbf{H}_m^2},\end{aligned}\tag{31}$$

where $\epsilon''(\omega)$ and $\mu''(\omega)$ are the permittivity (that includes conductivity losses) and permeability loss factors and ω_0 is the center angular frequency. The first expression on the RHS of Eq.(31) is an exact microscopic representation that relates to the macroscopic description. The second form is for dissipative media, averaged over a cycle, and we used the approximate expression for the stored energy $u_{eff}(\mathbf{r},t)=\Re[\frac{d(\omega\epsilon)}{d\omega}(\omega_0)]\overline{\mathbf{E}_p^2}(\mathbf{r},t)+\Re[\frac{d(\omega\mu)}{d\omega}(\omega_0)]\overline{\mathbf{H}_m^2}(\mathbf{r},t)$, which satisfies the approximate equation $\partial u_{eff}/\partial t+\nabla\cdot(\mathbf{E}_p\times\mathbf{H}_m)=-2\omega_0\epsilon''(\omega_0)\overline{\mathbf{E}_p^2}-2\omega_0\mu''(\omega_0)\overline{\mathbf{H}_m^2}$ [34, 35]. $\overline{\mathbf{E}_p^2}$, $\overline{\mathbf{H}_m^2}$ are the magnitudes of the fields time-averaged over one cycle ($2\pi/\omega$). The entropy-density rate can be written by means of Eqs.(19), (24), (25), and (31)

$$\frac{ds_d(\mathbf{r},t)}{dt}+\nabla\cdot\mathbf{S}_{eh}+\nabla\cdot\mathbf{J}_q\approx-\frac{\mathbf{S}_{eh}\cdot\nabla T}{T}+\frac{\nabla T\cdot\nabla T}{T^2}+\frac{2\omega_0}{T}\left\{\epsilon''(\omega_0)\overline{\mathbf{E}_p^2}+\mu''(\omega_0)\overline{\mathbf{H}_m^2}\right\}.\tag{32}$$

We assume that changes in the internal energy are due to the applied field entering the system $-\nabla\cdot(\mathbf{E}\times\mathbf{H})$ and a heat entropy flux $\mathbf{J}_q=-k\nabla T/T$. The Thompson-effect-related entropy flux is $\mathbf{S}_{eh}(\mathbf{r},t)=(\mathbf{E}\times\mathbf{H}-\mathbf{E}_p\times\mathbf{H}_m)/T$ where a temperature gradient drives a current and the reverse effect. Equation (32) expresses the important result that for passive systems, when the time-harmonic fields are averaged

over a cycle, the imaginary part of the permittivity and permeability must always be positive. Therefore both energy conservation and entropy production produce this constraint.

3.3. Frequency-Domain, Fluctuation-Dissipation, Entropy Production, and Material Response

It is well-known that noise, friction losses, and electric and magnetic loss parameters can be expressed in terms of fluctuation-dissipation relations in the weak-field limit. We now study approximations to Eq.(19) and obtain simplified expressions in the frequency domain for entropy rate related fluctuation-dissipation relations.

An approximation to Eq.(20) is

$$\frac{dS(t)}{dt} = \frac{1}{k_B}\int_0^t Tr\left(\dot{s}(t)\mathcal{T}(t,\tau)\bar{\dot{s}}(\tau)\sigma(\tau)\right)d\tau. \tag{33}$$

The LHS of this equation is the total entropy production rate from dissipative processes and RHS are the fluctuations. Noting that $\dot{s} = \int d^3r((\dot{u} - \dot{\mathbf{p}}\cdot\mathbf{E}_p - \dot{\mathbf{m}}\cdot\mathbf{H}_m)/T)$ we see that Eq. (33) combines the fluctuation-dissipation properties of the various components of the entropy rates[36].

We can approximate the spectral form of Eq.(33) using a $i\omega$ Laplace transform

$$L[\frac{dS(t)}{dt}](\omega) \approx \frac{1}{k_B}Tr(L[\dot{s}\dot{s}]L[\sigma]. \tag{34}$$

With this definition, analogous to the complex electromagnetic power for time-harmonic fields, the complex spectral entropy production rate has a real part that relates to dissipation and an imaginary part that relates to stored energy. In the next subsections we will summarize the contributions to the entropy production rate in Eq.(34) due to electric, magnetic, and internal-energy contributions individually and then combine the terms.

3.3..1 Linear Driving by an Electric Field

To begin, let us first review the fluctuation-dissipation theorem for electric polarization that relates to one term in Eq.(33) or (34). In order to study this, we need an expression for the pulse-response function. For electric-polarization response in a linear, time-harmonic, high-temperature approximation we obtain from Eq.(21)

$$\frac{d\mathbf{P}(r,t)}{dt} = \int d^3r'\int_0^t \frac{Tr(\dot{\mathbf{p}}(r)\mathcal{T}(t,\tau)\dot{\mathbf{p}}(r')\sigma(\tau))\cdot\mathbf{E}_p(r',\tau)}{k_BT}d\tau \tag{35}$$

$$\rightarrow \int d^3r'\int_0^\infty \frac{<\dot{\mathbf{p}}(r)\dot{\mathbf{p}}(r')>_0}{k_BT}\cdot\mathbf{E}_p(r',\tau)d\tau. \tag{36}$$

From this equation we can identify the response tensor $\overleftrightarrow{f}_e$ for linear response as $\overleftrightarrow{f}_e\ (r,t) = \int d^3r' < \mathbf{P}(r,0)\dot{\mathbf{P}}(r',t) >_0 /k_BT \approx -Vd/dt < \mathbf{P}(0)\mathbf{P}(t) >_0 /k_BT$ and V is volume. The density function, σ_0, used to calculate these expectations are for equilibrium, which does not contain the effects of any time dependence in the fields.

In the case of linear driving by an electric field without spatial correlations, the fluctuation-dissipation relationship can be obtained from Eq.(34) for a linear, time-harmonic response,

$$\overleftrightarrow{\chi}_e''(r,\omega) = \int_0^\infty \overleftrightarrow{f}_e(r,t)\sin(\omega t)dt = -\frac{1}{k_B T}\int_0^\infty d\tau \int d^3r' \frac{d}{d\tau}(<\mathbf{P}(r,0)\mathbf{P}(r',\tau)>_0)\sin(\omega\tau)d\tau$$
$$= \frac{\omega}{2k_B T}\int_{-\infty}^\infty d\tau \int d^3r' <\mathbf{P}(r,0)\mathbf{P}(r',\tau)>_0 \cos(\omega\tau)d\tau. \tag{37}$$

Eq.(37) is a well-known fluctuation-dissipation relationship, independent of the applied field. Physically this is interpreted as follows: the random, microscopic electric fields in a polarizable, lossy media produce fluctuations in the polarization and the resulting loss. Due to the external electric field driving force, this system is in equilibrium or a weak nonequilibrium state. In addition to Eq. (37), one could also write a relation between fluctuations and the real part of the susceptibility that relates to the stored energy. Comparing Eq.(37) to Eq.(34), the electric-field contribution to the spectral-entropy production rate is $\Re\,\widetilde{\Sigma}_e(\omega) = (\sigma_c + \omega\chi_e'')|\mathbf{E}_p|^2|/2T$.

3.3..2 Linear Driving by a Magnetic Field

Magnetization fluctuations are important in the magnetic-storage technology with respect to signal-to-noise ratio limitations [37]. This noise can also be modeled by fluctuation-dissipation relations for magnetic response. The linear fluctuation-dissipation relation for the magnetic loss component can be derived similarly to the electric field

$$\overleftrightarrow{\chi}_m''(\omega) = \int_0^\infty \overleftrightarrow{f}_m(\tau)\sin(\omega\tau)d\tau = -\frac{V\mu_0}{k_B T}\int_0^\infty \frac{d}{d\tau}(<\mathbf{m}(0)\mathbf{m}(\tau)>_0)\sin(\omega\tau)d\tau$$
$$= \frac{\omega V\mu_0}{2k_B T}\int_{-\infty}^\infty <\mathbf{m}(0)\mathbf{m}(\tau)>_0 \cos(\omega\tau)d\tau = \frac{\omega V\mu_0}{4k_B T} <\mathbf{m}^2>_0, \tag{38}$$

where $<\mathbf{m}^2>_0$ are the magnetic-moment fluctuations per Hertz. Therefore, the magnetic-field contribution to the entropy production rate is $\Re\,\widetilde{\Sigma}_h(\omega) = \omega\chi_h''|\mathbf{H}_m|^2|/2T$.

3.3..3 Internal-Energy Contributions

In this subsection we assume that the system allows heat exchange with the surroundings. We know the change in the internal energy density is $\partial U/\partial t = -\nabla\cdot\mathbf{Q} - \nabla\cdot(\mathbf{E}\times\mathbf{H})$ and originates from the applied fields and heat energy flowing in or out through the bounding surfaces.

We can write the internal-energy entropy production rate as

$$\Sigma_h(\mathbf{r},t) = \nabla\cdot\int_0^t Tr\left(\frac{\mathbf{j}_u(\mathbf{r})\mathcal{T}(t,\tau)(1-P(\tau))\mathbf{j}_u(\mathbf{r}')}{k_B T}\sigma(\tau)\right)\cdot\left(\frac{\nabla T(\mathbf{r}',\tau)}{T^2(\mathbf{r}',\tau)}\right)d\tau, \tag{39}$$

where $\nabla\cdot(\mathbf{j}_u(\mathbf{r})) = -\dot{u}(\mathbf{r})$.

The thermal conductivity transport coefficient can be identified by comparison of Eq.(39) to the time-dependent heat transfer equation: $\rho_d C_v\partial T/\partial t = \nabla\cdot\overleftrightarrow{\kappa}\cdot\nabla T$

$$\overleftrightarrow{\kappa} = \frac{1}{k_B T^2}\int_0^\infty d\tau\int d^3r' <\mathbf{j}_u(\mathbf{r})\mathcal{T}(t,\tau)(1-P(\tau))\mathbf{j}_u(\mathbf{r}')>. \tag{40}$$

Also an inspection of Eq.(39) indicates that the heat capacity per unit volume C_v times the density ρ_d is related to the fluctuations in the internal energy[21]

$$\rho_d C_v T = \int d^3r' \frac{< u(\mathbf{r})u(\mathbf{r}') >}{k_B T}, \tag{41}$$

where ρ_d is the density. Therefore the internal-entropy-production, by inspection of Eqs. (39) and (40), is approximated as

$$\Re\Sigma_{d(u)} = \kappa \frac{\nabla T \cdot \nabla T}{T^2}. \tag{42}$$

If we combine all our entropy-production contributions for time-harmonic fields and non-uniform temperature we have

$$\Re \widetilde{\Sigma}_d(\omega) = \frac{1}{2T}\left[(\sigma_c + \omega\chi_e'')|\mathbf{E}|^2 + \omega\chi_m''|\mathbf{H}|^2 + 2\kappa\frac{\nabla T \cdot \nabla T}{T} - \frac{\mathbf{S}_{eh\omega}\cdot\nabla T}{T}\right]. \tag{43}$$

We used $\mathbf{S}_{eh\omega}(\mathbf{r},t) = (1/2)\Re(\mathbf{E}\times\mathbf{H}^* - \mathbf{E}_p \times \mathbf{H}_m^*)/T$. Of course there are other contributions from the field interactions in the internal energy that we neglected and the cross-correlation terms.

3.3..4 Nyquist Noise

Noise in electromagnetic systems is usually separated into contributions from specific sources such as Johnson-Nyquist thermal noise, magnetic polarization noise, flicker noise, and shot noise. Shot noise may originate from variations in charge and mass flux away from equilibrium. The Nyquist formulation of Johnson noise is valid for stationary processes. Equation (34) indicates that fluctuations in entropy rate relate to the total contributions of all the noise sources. In the literature a number of researchers have conjectured that the commonly used noise definitions may be inadequate at low temperatures and high frequencies, and also for nonequilibrium behavior [38]. At millimeter-wave frequencies and above, the nonlinearity in Planck's law at low temperature cannot be neglected and contributions of quantum fluctuations can be important and $k_B T$ must be replaced by the effective energy E_β [39]. The thermal noise process in equilibrium without a macroscopic driving voltage is usually modeled using the principle of detailed balance where the electromagnetic power absorbed by a resistor is balanced by the emission of electromagnetic energy by the resistor. In this view, random fluctuations in the velocity of charges in a resistor produces a zero mean voltage, but nonzero fluctuations in the voltage in a circuit. This produces a net power flow in the circuit, but at the same time absorption of the electromagnetic fields by the resistor returns an equal amount of electromagnetic power to heat. The exchange of heat and electrical energy or the fluctuations in brownian motion require a reservoir to conserve energy[40]. The approach in this paper is to understand the noise process in terms of the concept of entropy production rate and entropy fluctuations.

We can apply the principle of detailed balance for a system in equilibrium using entropy production rate to obtain the Nyquist relations or black-body radiation. An argument based on entropy production rate follows the same lines of reasoning as Nyquist followed using power fluctuations. The local entropy

production rate due to random currents in the resistor, by Eq. (18) has a zero mean since the charged-particle motion in forward and time-reversed paths compensate. At equilibrium, the entropy production rate fluxes that result from the transmitted and absorbed electromagnetic waves in the circuit exactly balance. However, the fluctuations in the microscopic entropy rate is nonzero, by Eq. (19), just as $< v^2 > \neq 0$ in Nyquist's approach.

In order to derive the Nyquist relation from Eq. (19) we begin with a isolated system that is composed of a waveguide terminated at both ends with a resistance R. If we use Eq. (19) in the linear approximation, the microscopic entropy production rate that is due to random fluctuating charge densities $\mathbf{j}_m$ is $\dot{s} \rightarrow \int d^3r \mathbf{j} \cdot \mathbf{E}/T \rightarrow (1/2) I_0 v/T$, where I_0 is a bias current. Also the entropy production rate is $\widetilde{\Sigma} = (1/2) I_0^2 R$. For a stationary process, and use of the Wiener-Khinchine theorem, this reduces to the Nyquist relation $4k_B T R \Delta f =< vv^* >$, which can be expressed in terms of the entropy production rate in the waveguide resistors as $dS/dt \rightarrow k_B \Delta f =< v^2 > /4RT$.

Nonequilbrium, nonstationary noise problems have not been extensively studied. Equation (19) can, in principle, be used to form noise relations for nonequilibrium systems. If we express the entropy production rate in terms of the microwave power: $\mathcal{P}/T$

$$\frac{dS}{dt} = \frac{1}{T} Tr \left(\int_0^t \left(\frac{\mathcal{P}(t)\mathcal{P}(\tau)}{k_B T} \right) \sigma(\tau) d\tau \right) \rightarrow Tr \left(\int_0^t f_{pp}(t-\tau) \sigma(\tau) d\tau \right), \tag{44}$$

where we assumed that the power-power function $f_{pp} = V k_B (\mathcal{P}(t)\mathcal{P}(\tau))/(k_B T)$ depends only on $t - \tau$. By taking a Laplace transform of Eq.(44) we obtain a generalization of the Nyquist formula in terms of the density function $\widetilde{\sigma}\ (\omega)$ when the noise in the impedance Z is not necessarily white $Tr(f_{pp}(\omega)\ \widetilde{\sigma}\ (\omega))$. In the special case of Johnson noise

$$2k_B T \Delta f = \frac{< v(\omega) v^*(\omega) >}{Z(\omega) + Z^*(\omega)}. \tag{45}$$

Equation (19) is completely general, so it can be used as a basis to model nonequilibrium noise problems.

3.4. Boltzmann's Constant

From Eq. (19) we have an exact relation for Boltzmann's constant in terms of the total entropy production ratedS/dt and the fluctuations in microscopic entropy production

$$k_B = \frac{\int_0^t Tr \left(\dot{s}(t) \mathcal{T}(t,\tau)(1 - P(\tau)) \bar{\dot{s}}(\tau) \sigma(\tau) \right) d\tau}{dS(t)/dt - dS(0)/dt}. \tag{46}$$

In this context, entropy production rate is a natural way to estimate Boltzmann's constant. Note that the entropy production rate could be from mechanical, electrical, or hydrodynamic sources. This expression reverts to the standard Johnson noise relation for stationary processes. However, the expression is much more general in that any system that has entropy production rate could be used to obtain Boltzmann's constant.

3.5. Applications to Cavity Resonance and Permittivity, Permeability Constraints

A cavity resonator may be considered as an example of a nonequilibrium system in the sense that it requires applied forces to maintain the resonant state. The Q of a cavity is defined in terms of the resonant frequency ω_0 and the internal energy $\mathcal{U}(t)$ by the relation $d\mathcal{U}/dt = -(\omega_0/Q)\mathcal{U}$. Therefore the associated total entropy production rate is $(\omega_0/QT)\mathcal{U}$. For time-harmonic fields, just as the total power is complex, the entropy rate is also complex ($\Sigma = \Sigma_R + i\Sigma_I$) where the real part relating to dissipation and the imaginary part relates to reactive and stored energy. Near the resonance frequency, a cavity resonator achieves maximum dissipation and maximum stored energy, and therefore achieves a maximum in entropy production. Since the reactance goes through zero near resonance the reactive part of the time-harmonic entropy rate goes through zero. Therefore if we treat a resonator as a nonequilibrium system, it attains maximum-entropy production rate through resistive losses and achieves minimum reactive entropy. This correlates with a driven, organized state. Of course not all nonequilibrium states maintain maximum-entropy production.

Recently, in the metamaterials literature it has been conjectured that the positivity of the loss factors in the permittivity and permeability may not hold. It must be noted that the non-vacuum permittivity and permeability is only defined for frequencies where the wavelength in the material is significantly larger than any inclusion length scale in the system being analyzed. If we use the fluctuation-dissipation relations in Eqs. (37), (38), and (43) [41, 42], for the susceptibility-response averaged over a complete cycle, we have a simple proof of the positivity requirement for ϵ'' or μ'', that is consistent with both conservation of energy and the positivity of the net entropy production rate in an isolated system. Therefore if either ϵ'' or μ'' were negative for a thermally closed, passive system, when averaged over a cycle, then both the fluctuation-dissipation theorem would fail and the second law of thermodynamics would be violated. The origin of this type of behavior most likely originates from a heterogeneous material that does not satisfy the basic averaging requirements to define a homogeneous permittivity or permeability.

4. Conclusions

We studied entropy evolution in dissipative electromagnetic systems at either electrical and thermal equilibrium and away from equilibrium. In a systematic development, we began by developing an exact entropy fluctuation-dissipation relation that we used to study the entropy production. In the frequency domain, approximations were used to obtain expressions for the electrical properties of materials, noise, and power. The entropy-based fluctuation-dissipation relations combine effects from many sources of dissipation. We used the theory to study entropy in waveguide systems, noise, Boltzmann's constant, cavity resonances, permittivity, and entropy flow in electromagnetic systems.

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